Development of a Novel Method for Airborne Dust Reduction and Bioaerosol Deactivation Using Engineered Water Nanostructures (EWNS)

A Thesis Submitted to the College of Graduate and Postdoctoral Studies in partial fulfillment of the requirements for the degree of

Master of Science

in the Department of Chemical and Biological Engineering

University of Saskatchewan

By

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Abstract

Airborne pathogens and dust particulates are associated with the infectious and respiratory diseases that increase morbidity and mortality in livestock operations.

An environment-friendly, non-toxic, and inexpensive technique was developed for airborne bacterial inactivation in livestock operations. This technique is based on electrospray (ES) which converts water into highly charged nano-scale water droplets or engineered water nanostructures (EWNS). The EWNS has some unique physicochemical characteristics that may be important for dust reduction and bacteria inactivation. The nano-scale water droplets have high mobility, charged density, long lifetime, and contain reactive oxygen species (ROS), all of which would be beneficial for airborne dust reduction and bacterial inactivation.

To date, there are no studies on the effectiveness of EWNS nanoscale droplets on airborne dust reduction and bioaerosol inactivation in livestock buildings. Livestock buildings have high ventilation rates, large variation of climatic parameters, and high concentration of airborne dust and bioaerosols in comparison to residential buildings. Therefore, the objective of this study was to design and optimize a laboratory-scale electro-spraying system for generating engineered water nanostructures (EWNS) and to test the airborne dust reduction and bioaerosol inactivation of EWNS.

For airborne dust reduction, both poultry and swine dust, with size less than 50 μm were used to test the dust reduction of EWNS. The swine dust concentrations for both control and treatment trials were 14.3 mg/m^3 at 5 air changes per hour (*ACH*) and 12.5 mg/m^3 at 15 air changes per hour (*ACH*), respectively. The poultry dust concentration was 13.5 mg/m^3 at 15 air changes per hour (*ACH*). The experimental results revealed that the highest dust reduction percentage was 72.9% and 83.6%, at 15 air changes per hour (*ACH*) for swine and poultry dust, respectively. Moreover, the liquid consumption of the newly designed EWNS generator was a

maximum of 1.92 ml/h, which is much less than most of the studies for the applications of wet electrostatic scrubbers (WES) for air quality control in livestock barns.

For the bioaerosol inactivation experiment, *Escherichia coli* W3110 was used as a representative strain for the bioaerosol generation. The airborne *E. coli* concentrations for both control and treatment trials were $(2.0 \pm 0.6) \times 10^4$ CFU/m^3 and $(3.1 \pm 0.1) \times 10^3 \pm$ CFU/m^3 at ventilation rates of 7 and 15 air changes per hour (*ACH*), respectively. The experimental results revealed that sprayed liquid at pH 7 had the highest inactivation of 69% at 7 air changes per hour (*ACH*), and 37% at 15 air changes per hour (*ACH*), respectively. Moreover, the sprayed liquid consumption of the newly designed EWNS generator was only 480 µl/h and EWNS particle number concentration can up to 120,000 #/ cm^3 in a 250-L experimental chamber.

Overall, the results from both airborne livestock fine dust reduction and bioaerosol inactivation indicated that the EWNS have a set of unique physicochemical properties and this newly designed EWNS generator is effective at reducing airborne dust concentrations and inactivating *E. coli*.

Acknowledgements

First of all, I would like to express my gratitude to my supervisors, Dr. Lifeng Zhang and Dr. Shelley Kirychuk, for providing me the chance to be a member of the research group and on top of that for their continuous support and guidance in overcoming numerous obstacles I have been facing through this challenging research.

I am also grateful to Dr. Huiqing Guo and Dr. Bernardo Predicala for their feedback, support and cooperation in this work which helped me to get results of better quality.

Also, I would like to thank my committee member, Dr. Catherine Niu from the Department of Chemical and Biological Engineering for providing me with valuable advice and suggestions during the research.

I also acknowledge the financial support from the Agriculture Development Fund (ADF) from the Ministry of Agriculture of Saskatchewan, and Agrivita Canada Inc.

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Nomenclature

Symbol	Definition
a	Measured radius of spread droplet (nm)
ACH	Air changes per hour (ACH)
C_i	Average dust mass concentration of control trials (mg m^{-3})
C_e	Average dust mass concentration of treatment trials (mg m^{-3})
C_{TB}	Total bacteria concentration (CFU m^{-3})
C_{Bi}	Cocentration of airbrone bacteria in size interval i (CFU m^{-3})
C_{TBC}	Total airborne bacteria concentration in control trial (CFU m^{-3})
C_{TBT}	Total airborne bacteria concentration in treatment trial (CFU m^{-3})
d	Equivalent diameter of spread droplet (nm)
d_P	Particle diameter (nm)
D	Diameter of sampling hole on counter electrode plate (cm)
h	Height of the deposited droplets (nm)
K	Electrical conductivity (S m^{-1})
L	Electrode to counter electrode plate distance (cm)
Q	Flow rate of spread liquid (µl min^{-1})
V	Applied voltage between electrode plates (kV)

Greek letters

Symbol	Definition
σ	Surface tension (N/m)
ε_0	Vacuum permittivity (C^2 ($N\ m^2$) $^{-1}$)
µ	Dynamic viscosity (kg/(m s))
η	Reduction percent (%)

ρ	Fluid density (kg/m^3)
β	Relative permittivity
α_ρ, α_μ	Non-dimensional parameter

Chapter 1 - Introduction

Dust and bioaerosol concentrations have the potential to impact the health of animals and workers in animal confinement facilities [1, 2]. A high prevalence of respiratory, eye and skin conditions have been reported in animal confinement facility workers [3,4]. Reducing dust and bioaerosol concentrations in confined agricultural operations could improve worker health. Conventional methods such as ventilation and filtration are less efficient at removing particulates, in particular, dust and bioaerosol of submicron size, as compared to nano electrospray (ES) [5,6]. The nano-ES can generate nano-scaled water droplets (EWNS) with high charge density and mobility that will increase coagulation efficiency between droplets and dust particles [7] and thereby improve removal from the airspace. In addition, the EWNS contains reactive oxygen species (ROS) that can deactivate bacteria [6]. Overall, this method is considered more environment-friendly and cost effective than the traditional methods mentioned above.

Electrospray (ES) has been used in various fields, such as, electrospray ionization (ESI) in mass spectrometry, coating conductive materials, and generating monodisperse aerosols [5]. As seen in Figure 2.1, a typical electrospray unit includes a power supply, a syringe pump, a needle, and a counter electrode. The high-voltage power supply is used to generate a high electric field between the needle and the counter electrode. The syringe pump is used to control the spray liquid flow rate. It is convenient to divide the process of electrospray into two stages: spray mode formation, and droplet formation with shrinkage. The liquid delivered to the tip of the needle experiences the electric potential which can be either positive or negative. Assuming a positive potential, the liquid with positive charges will accumulate near the tip of the needle. When the imposed electrical field is high enough, and the electrostatic force is greater than the surface tension of the liquid, the spray mode will start to form [8]. In stage II, owing to the

electrostatic repulsion and the Rayleigh limit, droplets will start to split into fine droplets [9-10]. The diameter of the droplets formed is determined by applied voltage, spray liquid properties, needle diameter, and liquid flow rate.

1.1 Project motivation and knowledge gap

Despite considerable progress made by various researchers through the past few decades, the effects of nano-scale water droplets generated through electrospray system on airborne dust removal and bioaerosol inactivation are not clear. In addition, there is no study of the use of nano scale water droplets to reduce livestock barn dusts and test its effectiveness for airborne bacteria inactivation.

These nano-scale water droplets have some unique physicochemical properties that make them a good alternative method for air quality control. Moreover, the effects of nano-scale water droplets on livestock barn dust reduction and bioaerosol inactivation are not known. Our previous work investigated the total current and deposition area of sprayed water droplets by varying the chemical and physical properties of the sprayed water droplets through changing the pH, electrical conductivity, surface tension, density, and dielectric constant was published in Results in Engineering [10].

Two main chemical reactions are associated with changing pH of the spray liquid. The concentration of hydroxide ions will be increased when adding alkaline chemicals into the water; whereas adding acidic chemicals to the water will increase concentration of hydrogen ions. As demonstrated in the literature review section, the charge density and reactive oxygen species (ROS) of the sprayed water droplets are the key factors for airborne dust reduction and bacteria inactivation. Varying the total ions concentration, and in particular hydroxide ion that contributes the most as the source of hydroxyl radicals among all the ROS for the bacteria inactivation. Thus, alkaline addition might increase the charge density and generate more

hydroxyl radicals resulting in improved airborne dust reduction and bacteria inactivation. The reverse osmosis water with pH of 7, sodium hydroxide and water mixtures with pH of 9 and 12, and 0.9% mass concentration saline will be tested.

There is also a need to investigate a broader range of operating conditions on EWNS droplet size distribution. Such as applied voltage, polarity of charged water droplet, and sprayed liquid flow rate, to evaluate the effects of different operating parameters on the EWNS droplet size. The knowledge from this work provides the foundation for pilot and commercial scale EWNS design and operation. The most optimized operating conditions from a previous study of "Investigating Efficiency of Engineered Water Nanostructures Generated via Electrospray Technique to Deactivate Surface Microbes in Livestock Barns" were used to test airborne bacteria inactivation [11].

Overall, the research hypotheses are:

1. The nano scale droplet generated through electrospray is an effective method in reducing livestock barn dusts and airborne bacteria at high concentration conditions.
2. Different applied voltage and charge polarity of nano scale droplet can affect livestock barn dust reduction.
3. Higher pH solution is more effective in dust reduction and bacteria inactivation compared to neutral pH.
4. Higher electrical conductivity solution generates the droplet with longer lifetime.

1.2 Research objectives

The objective of the work was to systematically investigate the effects of nano-scale water droplets on livestock barn dust reduction and bacteria inactivation. The dust reduction was tested over a wide range of voltage (-5.5, -6.5, +5.5 and +6.5 kV), three sets of spray liquid flow rates (1, 2 and 4 µl/min), three different distances of the electrode plate (2, 3 and 4 cm),

and various levels of pH (7, 9 and 12), to determine the most optimized operating condition with the highest reduction percent. For the airborne bacteria inactivation experiment, the most optimized operating conditions from a previous study using EWNS on surface bacteria deactivation including applied voltage of -5.5 kV, sprayed liquid at a flow rate of 1 µl/min, and electrode distance of 2 cm were employed and tested with three different spray liquids (pH 7, pH 12 RO water, and 0.9% saline) [11].

1.3 Organization of thesis

The work presented in this thesis resulted in two manuscripts. The thesis is written in the manuscript-based style and organized into five chapters. The introduction is presented in Chapter 1, along with knowledge gap, objectives, and thesis organization. In Chapter 2, the literature review provides background on the principles of an electrospray system, mechanism of dust collection, ROS generation, and inactivation on bacteria, as well as spray modes of electrospray, scaling laws to predict liquid droplet size, and toxicological evaluation of EWNS. Chapter 3 includes the first manuscript with focus on determining the effects of different operating conditions for EWNS on livestock fine dust reduction including the applied polarity and voltage, sprayed liquid flow rate, electrode distance, and pH of spray liquid. Chapter 4 contains the results of airborne *E. coli* inactivation using the EWNS method and a comparison between EWNS and non-thermal plasma on *E. coli* deactivation efficacy. Chapter 5 presents a summary of results, conclusions, and recommendations.

Chapter 2 – Literature review

In the following sections, the principle and spray modes of electrospray, scaling laws to predict average droplet size and total current, alternative technologies for dust reduction and microbial deactivation, mechanism of dust collection and bacteria inactivation, and toxicological evaluation of EWNS are provided.

2.1 Electrospray principle

Electrospray is an electrohydrodynamic technique similar to electrospinning. Both techniques are governed by a similar principle and typically use identical apparatus as shown in Figure 2.1. The typical apparatus includes a high voltage power supply, a syringe filled with a solution and equipped with a metallic capillary as electrode, a syringe pump controlling the sprayed liquid flow rate, and a counter electrode plate. When the electric potential is applied between the electrode and counter electrode, a stable Taylor cone will form, which is stabilized by the liquid surface tension, electrostatic force, and gravity [12]. Owing to the electrostatic repulsion and Rayleigh effect, droplets generated from the Taylor cone are highly unstable, and they will start to split into fine droplets [9-10].

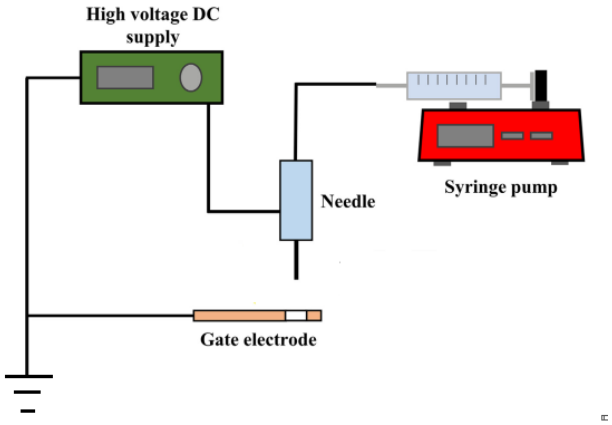

Figure 2.1 The basic apparatus of electrospray from Lee and Kim [13] (Page 192). [With permission from Elsevier].

2.2 Spray modes of electrospray

The spray mode differs between fields, which depend on the application. For instance, the cone-jet mode is used to generate EWNS for airborne pathogen inactivation as well as in electrospray ionization (ESI) in mass spectrometry for biological sample analysis. In contrast, a microdripping mode has been used to prepare monodispersed microdroplets for removal of fine dust particles smaller than 10 μm [13]. The spray modes are influenced by many parameters, such as, spray liquid properties, liquid flow rate, applied voltage, electrode-to-counter-electrode distance, and spray needle size. Figure 2.2 shows the common spray modes of electrospray.

The types of the spray modes can be classified based on the geometrical form of the liquid at the tip of the nozzle and the mechanism of the disintegration of the jet into droplets.

In general, the spray modes can be categorized into two groups, the first group includes dripping mode, microdripping mode, spindle mode, multispindle mode and ramified-meniscus mode. In the first group, only fragments of the liquid are ejected from the nozzle, there is no jet formed. The second group includes cone-jet mode, precession mode, oscillating-jet mode, multijet mode, and ramified-jet mode. In the second group, the liquid is in the form of a long continuous jet, which disintegrates into droplets only in some distance, usually a few millimeters from the needle tip [14]. In the following section, the common spray modes will be discussed in greater detail.

2.2.1 Dripping mode

For the dripping mode, there is no significant difference between **dripping mode** that is caused by electrospray and **dripping** that is caused by gravitational force only. For **dripping mode** formation, when a voltage is applied, the droplets are formed as regular shapes detaching

from the nozzle as the weight of the droplet and the electric force overcomes the surface tension of the liquid [14].

2.2.2 Microdripping mode

For the microdripping mode, a liquid at the outlet of a nozzle forms a stable meniscus. At the end of the meniscus, droplets are formed, in which the sizes are much smaller than the nozzle diameter. The droplet detaching from the meniscus does not undergo further fission. Meanwhile, this mode occurs only at low liquid flow rates. Comparing to the dripping mode, the meniscus of the microdripping mode does not contract after droplet detachment [14].

2.2.3 Spindle mode

For the spindle mode, the thick jet-like meniscus of the liquid elongates in the direction of an electric field. Droplets detach as a vast spindle-like fragment of liquid. After the detachment, the spindle changes to several smaller droplets in different sizes, which disperse off the nozzle axis. After the spindle detachment, the meniscus contracts to its original shape and a new thick jet starts to form [14].

2.2.4 Cone-jet mode

For the cone-jet mode, the liquid forms a regular, axisymmetric cone with a thin jet (<100 μm in diameter) at its apex. The cone can take three different forms: with linear sides, convex and concave [15]. The jet flows along the nozzle axis or deflects from it only at a small angle (<10°). The jet at its end undergoes instabilities, which include varicose and kink [14, 16].

2.2.5 Multijet mode

For the multijet mode, the meniscus tends to be flat with small cones at distinct points at the circumference of the nozzle, and the fine jets of liquid are ejected. The jet disintegrates due to kink instabilities, into small droplets forming a fine mist around the nozzle [14].

2.2.6 The significance of cone-jet mode for indoor air treatment

It is significant to ensure the electrospray is operated under the cone-jet mode in this project. The advantage of using the cone-jet mode for airborne bacteria inactivation and dust reduction includes its steady production of self-dispersed unipolar droplets, which carry charges closer to the Rayleigh limit than the droplets generated from other types of spray modes. The closer to the Rayleigh limit, the higher charge density of droplets will be formed, which will increase the coagulation efficiency between droplets and dust particles as well as increase the electrical mobility of droplets. In addition, a stable cone-jet mode will produce unimodal-sized droplets, which is significant for size control of nano-scaled droplets [5]. The non-unimodal distribution will not generate a binomial droplet size distribution, and it will have droplets larger than 100 nm that could affect the airborne bacteria inactivation, as droplets smaller than 100 nm are more efficient for airborne bacteria inactivation [6].

Thus, the cone-jet mode is the best mode for producing the spray type required for EWNS. To do this, the differences of shape for each spray mode need to be identified in order to distinguish the cone-jet mode from other spray modes.

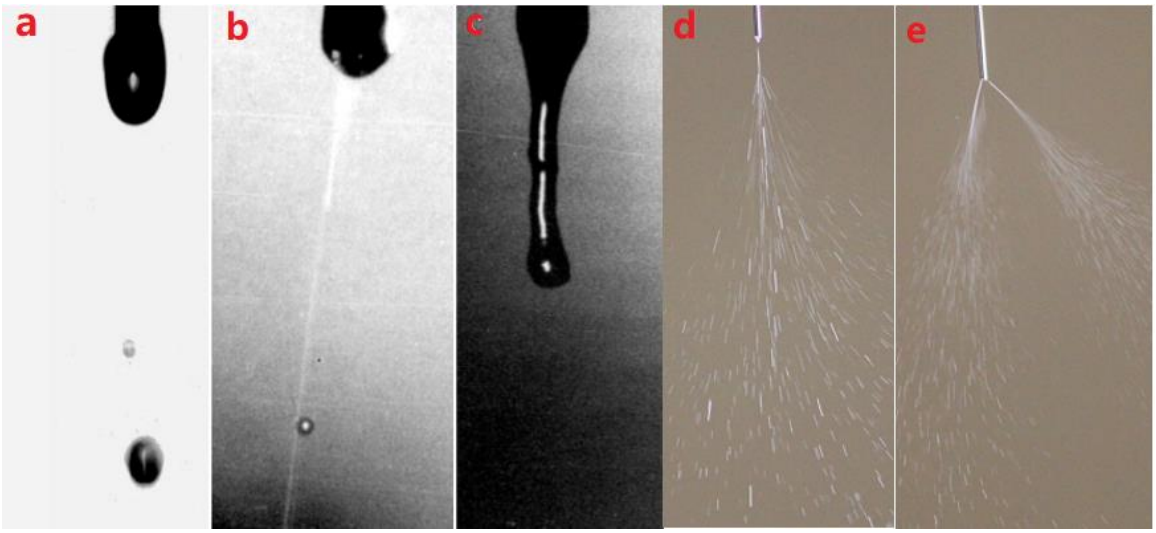

Figure 2.2 The common spray modes of electrospray (a) dripping mode (b) microdripping mode (c) spindle mode (d) cone-jet mode (e) multijet mode from Jaworek et al. and Xie et al. [14, 17] (Page 47 and 33). [With permission fromm Springer Nature and Elsevier].

2.3 Scaling laws

The scaling law of electrospray is a functional relationship for predicting droplet size and currents. The law is based on experimental parameters such as, spray liquid properties, spray mode, applied voltage, separation distance of electrode and nozzle diameter. Different scaling laws have been described in the literature and some scaling laws for predicting droplet size and total current are based on different operational conditions and spray modes [17].

2.3.1 Scaling law of total current and mean droplet size for Cone-jet mode

Because of the spray liquid properties used in this project, there is only one asymptotic scale, out of six, that can be used. The dominance of inertia and electrostatic suction scaling was originally proposed by Ganan-Calvo's group [16] and Hartman [18] without any constraints. In the publication of Ganan-Calvo (2004), the following constraints need to be satisfied to use the dominance of inertia and electrostatic suction scaling :

$$\alpha_\rho \gg {\alpha_\mu}^{1/4}\ , \quad \frac{\alpha_\rho}{\beta-1} \gg 1. \tag{2.1}$$

Where, α_ρ and α_μ are two non-dimensional parameters defined below:

$$\alpha_\rho = \frac{\rho K Q}{\sigma \varepsilon_0}, \quad \alpha_\mu = \frac{K^2 \mu^3 Q}{{\varepsilon_0}^2 \sigma^3}, \tag{2.2}$$

If the constrains above are satisfied, the total current and droplet size can be determined by

$$I = (\sigma K Q)^{1/2} \qquad \text{and} \tag{2.3}$$

$$d = \left(\frac{\rho \varepsilon_0 Q^3}{\sigma K}\right)^{1/6} \tag{2.4}$$

where,

$\beta =$ Relative permittivity;

$\varepsilon_0 =$ Vaccum permitivity with a value of 8.854×10^{-12} F/m;

$K =$ Electrical conductivity;

$\mu =$ Dynamic viscosity;

$\rho =$ Sprayed liquid density;

$Q =$ Feed flow rate;

$\sigma =$ Liquid surface tension;

2.4 Alternative technologies for dust reduction and microbial inactivation

Current technologies for dust reduction in livestock buildings include ventilation control, dust abatement, and wet scrubbers. Ventilation control strategies involve controlling ventilation rate to emit pollutants to the outside of livestock buildings. Dust abatement strategies include the cleaning of surfaces and oil or water spraying to restrict dust emissions. Wet scrubber techniques use the combination of electrostatic precipitator and water spray to capture the airborne dust particles [19].

For shortcomings, the efficiency of ventilation control is dependent on the building configuration, outside temperature, and seasonal and climatic parameters that make it a key issue for pollutant removal [20]. The efficiency of oil or water spraying can only be effective at reducing large dust particles for several hours [21]. The oil spray also generated many more smaller particles that affect human respiratory capacities [22]. For the wet scrubber technique, the water consumptions were high, and a post-treatment of the aqueous solution was required [19, 23].

Current air disinfection technologies include upper-room UV-C irradiation (Wavelength of 254 nm), high-efficiency particulate air filtration (HEPA) and photocatalysis. UV-C light can destroy the structure of nucleic acids and inactivate microorganism. UV-C light disinfection is a chemical-free technology, and it has a wide range of applications, such as wastewater treatment and indoor air treatment [15, 24]. High efficiency particulate air filters are composed of fibers which are coated with silver and can kill bacteria. The HEPA filter has a large surface area that increases collection efficiency of dust particles and it can remove 99.97% of all particles greater than 0.3 microns [25]. Photocatalysis uses the reactive oxygen species generated by photocatalytic nanoparticles and ionized air, respectively to deactivate microorganisms. The photocatalytic nanoparticles need less activation energy to perform primary disinfectant reactions, which requires less energy [26, 27].

For shortcomings, UV-C irradiation requires the upper room installation of UV fixtures and it has potential health risks for workers, for instance, redness or ulceration of the skin under short-term exposures and premature aging of the skin as well as cancer under long-term exposures [28-29]. HEPA filters need large quantities of energy to circulate air through the filter, especially, when facing a high pressure drop condition, which together will increase the operational costs [30]. Meanwhile, HEPA filters are less efficient to remove volatile organic compounds (VOCs) and viruses as they are far smaller in size than what can be trapped [31].

Photocatalysis is restricted to surface deactivation and these commonly used photocatalytic nanomaterials, such as Ag or TiO_2, can not be used for airborne pathogen inactivation due to their toxicological effects, such as their abilities to deposit in all regions of the respiratory track and translocate to sensitive organs via the blood or lymph [32-33]. Meanwhile, TiO_2 nanoparticles have been demonstrated with its toxicity in human lung epithelium cells (A549) [34].

2.5 Current status using ES for bioaerosol inactivation and dust reduction

The airborne dust removal by using electrospray has been studied for decades. Recently, there are some studies using electrospray to deactivate pathogens on surface and in air.

For airborne dust control, previous study used a multi-nozzle electrospray system with precession mode and de-ionized water, doped with 0.1% by volume of non-ionic surfactant EXACT WETTER^{TM} to generate charged droplets. The generated size of droplets is between 60 and 110 μm. The process has been used for removing airborne smoke particles with efficiency as high as 80-90% for particles smaller than 1 μm [35]. Ehouarn et. al. used electrospray in a bipolar scrubber system, in which the charged dust particles met with oppositely charged water droplets, coagulated with each other, and deposited by gravity. The removal efficiency of particles with a mean diameter of 0.44 μm approached 99% [36].

For airborne bioaerosol inactivation, Georgios et al. used electrospray to generate EWNS, which demonstrated the ability to inactivate gram-negative (*S. marcescens*) and gram-positive bacteria (*S. aureus*) [6]. Under two different air changes per hour (*ACH*) conditions, the bacteria removal results are similar (50 vs. 40%) with 1.7 and 2.9 *ACH*, respectively [6]. More recently, the same research group has demonstrated that the EWNS are capable to inactivate influenza *H1N1/PR/8* virus in air and the viral concentration reduction efficiency was 94% compared to the controls [37].

2.6 Mechanism of dust collection and bacteria inactivation via EWNS

The spray of charged EWNS droplets is highly electrically charged and dispersed in the chamber and collects oppositely charged dust particles due to electrostatic Coulomb attraction forces between them, then deposited by the gravity [38- 39]

The EWNS contains reactive oxygen species (ROS), which are the key for bacterial inactivation. ROS are oxygen-containing radicals with one or more unpaired electrons. They are highly reactive and short-lived. ROS have been demonstrated as an oxidative stressor that cause damage to proteins [40] and DNA structures [41].

During the electrospray process, these ROS are generated. Such as, hydroxyl radicals ·OH, hydrogen peroxide H_2O_2, superoxide anion $O_2^{\cdot-}$ and ozone O_3. The following equations illustrate mechanisms of ROS production that are possibly responsible for bacteria deactivation [40].

$$H_2O \rightarrow \cdot\mathrm{OH} + H^+ + e^- \quad (2.5)$$

$$\cdot\mathrm{OH} \rightarrow 1/2O_2 + H^+ + e^- \quad (2.6)$$

$$\cdot\mathrm{OH} \rightarrow \cdot\mathrm{O} + H^+ + e^- \quad (2.7)$$

$$\cdot\mathrm{O} + O_2 \rightarrow O_3 \quad (2.8)$$

$$\cdot\mathrm{OH} + \cdot\mathrm{OH} \rightarrow H_2O_2 \quad (2.9)$$

$$\cdot\mathrm{OH} + O_3 \rightarrow HO_2^{\cdot} + O_2 \quad (2.10)$$

$$\cdot\mathrm{OH} + H_2O_2 \rightarrow HO_2^{\cdot} + H_2O \quad (2.11)$$

$$HO_2^{\cdot} \leftrightarrow O_2^{\cdot-} + H^+ \ (pK_a = 4.8) \quad (2.12)$$

Previous studies have demonstrated that the hydroxyl radical is the major molecule for *E. coli* inactivation [6, 42]. In particular, lipid membrane with polyunsaturated fatty acids is the primary target for hydroxyl radical. Hydroxyl radical leads to lipid peroxidation and thereby change the structure and activity of membrane proteins. As a result, lipid membrane is

decomposed to other products and cell will lose protection from membrane [43 - 44]. In Figure 2.3, the transmission electron microscopy (TEM) shows (D) control (unexposed to EWNS) bacteria and (E) treatment (exposed to EWNS for 90 minutes). It can be concluded that the treatment one (E) with damaged membrane structure is caused by ROS in the EWNS [6].

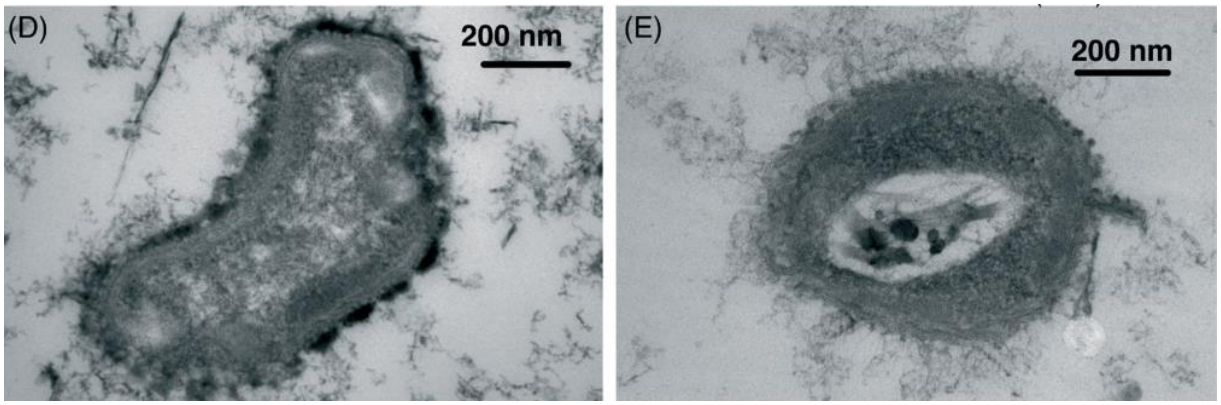

Figure 2.3 TEM images for control and treatment from Pyrgiotakis et al. [6] (Page 23). [With permission from Royal Society of Chemistry].

2.7 Toxicological evaluation of EWNS

A variety of nanomaterials can generate reactive oxygen species (ROS) under certain experimental conditions [37, 45 - 49]. ROS can lead to oxidative stress in cells [50], for this reason, the toxicological study needs to be performed to assess the potential health risk of EWNS.

From previous study, Georgios et al. used a bronchoalveolar (BALB)/c mouse model to evaluate the toxicological profile of inhaled EWNS [6]. There were three EWNS exposure levels, 10,000, 40,000 and 60,000 #/cm^3, respectively. After 24 hours post-inhalation exposure to EWNS, the albumin, hemoglobin, lactate dehydrogenase (LDH) [49], myeloperoxidase (MPO) [51], and inflammatory cells [52, 53] from BAL and nasal lavage (NL) were analyzed as indicators of injury and inflammation.

It can be seen in Figure 2.4 that there is no significant difference between the controls and EWNS-exposed animals for all indicators [6] of inflammation. Therefore, the EWNS contain ROS cannot cause any lung injury or inflammation and it was considered as a non-toxic disinfectant.

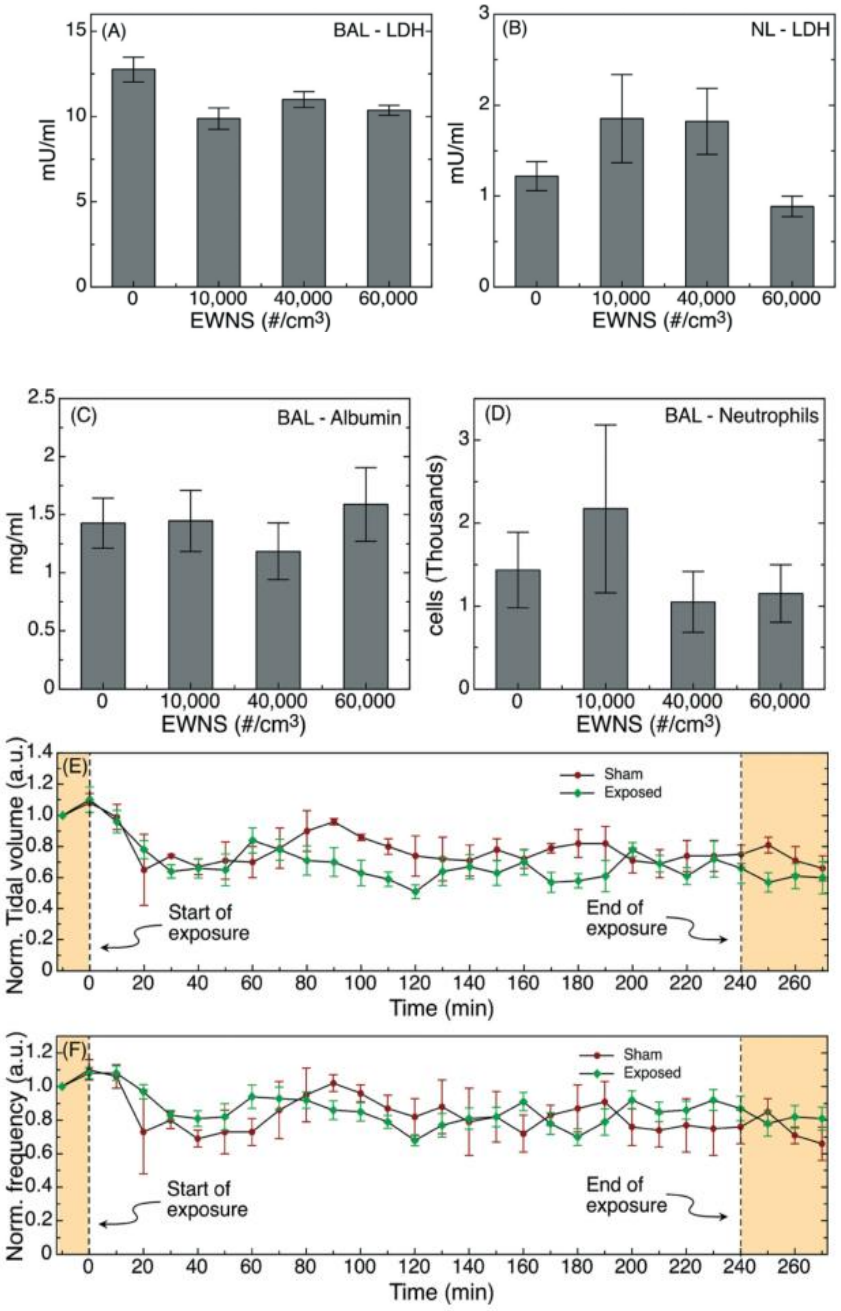

Figure 2.4 BAL and NL indicators of injury and inflammation post-exposure to EWNS. (A) BAL, LDH, (B) NL LDH, (C) BAL albumin and (D) BAL neutrophil numbers. (E) The normalized tidal volume and (F) normalized breathing frequency showed during and after the exposure no difference between exposure and controls from Pyrgiotakis et al. [6] (Page 24). [With permission from Royal Society of Chemistry].

2.8 Summary

Overall, the EWNS is an environment-friendly and non-toxic disinfectant and due to its unique physicochemical properties make it as a good alternative method for air quality control. There were lots of studies on using electrosprayed droplets with size greater than 1μm for airborne dust reduction and bacteria inactivation. However, there were fewer studies on using droplets with size less than 500 nm for air quality control and especially inside the livestock barns. Meanwhile, the water consumptions of previous studies are quite high that will consume lots of water and have potential risk of flooding in commercial livestock facilities. Therefore, in this project was aiming to design a new EWNS generator and optimize the operating parameters (i.e., applied voltage, electrode distance, flow rate) to reach the highest reduction percentage for both swine and poultry dust. Moreover, testing the most optimized operating condition from the previous project with different pH of liquids to determine bacteria inactivation efficacy at high level concentration of *E. coli* [11].

Chapter 3 – Experimental Studies on Removing Airborne Fine Dust Particles using Engineered Water Nanostructures Generated from an Electrospray

Contribution of the MSc student

Experiments were planned and performed by Eric Yang with the guidance provided by Drs. Lifeng Zhang and Shelley Kirychuk. Drs. Lifeng Zhang and Shelley Kirychuk supervised and provided consultation during the entire experimental period as well as thesis preparation. All the writing of the submitted manuscript was done by Eric Yang with Drs. Lifeng Zhang, Shelley Kirychuk, Huiqing Guo, and Bernardo Predicala providing editorial guidance regarding the style content of the paper.

Contribution of this chapter to the overall study

In this chapter, a newly designed EWNS generator was employed to test the swine and poultry dust (0.1 – 10 μm) reduction at two different ventilation rates of 5 and 15 air changes per hour (*ACH*). For the process of optimization, the operating parameters include applied voltage, polarity of charge, sprayed liquid flow rate, electrode distance and pH of sprayed liquid were optimized to find the operating condition with highest dust reduction and the most stable performance (small fluctuation). Meanwhile, the relationships between water droplet size and operating parameters were characterized by using an Atomic Force Microscope (AFM).

3.1 Abstract

A newly-designed prototype of engineered water nanostructures (EWNS) generator was developed and optimized under laboratory conditions for removal of airborne fine particle in livestock barns. Nanoscale water droplets with size less than 300 nm were generated through an electro nano-spray and tested at two ventilation rates (5 and 15 air changes per hour). Reverse osmosis water was used as the spray liquid and barn dust from swine and poultry operations was employed as airborne particles. This new generator design had a high EWNS generation rate (28,800 #/(cc min)) with water consumption of only 1.92 ml/h . The swine barn fine particle reduction percent was optimized by altering liquid flow rate, electrode distance, applied voltage, polarity of charges, sprayed liquid properties (i.e., pH). The optimized operating condition was tested for poultry barn fine dust removal at different pH levels. The experimental results revealed that the highest dust reduction at 15 air changes per hour (ACH) for swine and poultry barn fine dust is 72.9% and 83.6%, respectively.

3.2 Introduction

Concentrated animal confinement operations have led to a growing recognition of health and indoor air work environment issues [54]. Studies have shown that fine dust (PM10) in indoor air contributes to worker related health effects [3, 4, 55]. Moreover, in 2008, Environment Canada reported that agricultural operations represent approximately 18% of total PM10 open sources [56-57]. Thus, addressing particulate matter in concentrated animal confinement operations should assist in preventing occupational health and environmental effects.

Current technologies for controlling particulate matter include: (1) ventilation control, which consists of increased ventilation rate or filter-based air purification systems that require high energy to circulate air, high cost of filter replacement and frequent cleaning of the filtration

system [30, 58], as well as (2) electrostatic-based technologies, which include electrostatic precipitation (ESP) and wet electrostatic scrubbing (WES) [59-60]. Previous studies have shown the ESP has health and safety risks including the potential to increase indoor ozone concentration and fire hazard risks [61-62]. In addition, fine dust removal effectiveness has yet to be validated at the industrial scale [63-64]. In WES [38], the consumption of spray liquid was quite high (ml/min or l/min), making limited suitability for livestock barn air quality control, with potential for barn floor flooding and potential increase of the surface or airborne bacteria concentrations. Moreover, the average generated droplet sizes from WES studies were greater than 1 μm [23, 35, 62, 65-66], which makes the generated droplets have less charge density and mobility than the nanoscale droplets. In addition, the effectiveness of nanoscale water droplets for fine dust removal is not known.

A novel, nanotechnology-based, air purifying technique using Engineered Water Nano-structures (EWNS) generated from an electrospray can be an alternative method for airborne dust reduction in livestock buildings. Electrospraying is a widely used technology for aerosol generation with a controlled size range. The electrospraying process can be divided into two stages. In stage 1, a strong electric field is applied between a fine capillary containing a spray liquid and a counter electrode plate, causing the charges to accumulate near the tip of the capillary. When the imposed electrical field is sufficiently high and the electrostatic force is greater than the surface tension of the spray liquid, the spray mode will start to form [8]. In the second stage, owing to the electrostatic repulsion and the Rayleigh effect, droplets will start to split into fine droplets [9-10]. The size of the droplets is determined by the following parameters: applied voltage, spray liquid properties, the capillary diameter, and the spray liquid flow rate [67].

Versatile applications of the nanoscale water droplets have been demonstrated in various fields, such as bacteria inactivation, improvement of anti-pilling property of knitted

wool fabric, and textile odor removal [53, 68-69]. From a dust reduction perspective, EWNS are highly mobile and charged [6], which potentially increases the collection efficiency due to the smaller droplet size and high charge density [64, 70]. More importantly, this is an environment-friendly and toxicologically benign method, which does not pose any known potential health risks to either barn workers or animals [6]. However, there are no studies on the effectiveness of EWNS on airborne dust removal in livestock buildings.

Therefore, this study was aimed to develop and optimize a prototype electro nano-spray platform to achieve the highest dust reduction for both swine and poultry barn dust under controlled laboratory conditions.

3.3 Materials and methods

3.3.1 EWNS generator

In this study, a novel EWNS generator (Figure 3.1) was designed and optimized to test the performance of fine livestock dust removal in a controlled laboratory-scale setting. The EWNS generator has 8 electrospray units, with each unit consisting of a 30G stainless steel needle (Needle Metal Hub, Hamilton, USA) as an electrode. The new platform provides a stable water source through an 8-channel multi-syringe pump (NE-1800, NEW ERA, USA), which differs from the design from previous paper wherein Peltier-cooled electrode was used to condense the water vapor from surrounding air [6]. In such a design, the stability of flow rate will be affected by variation in relative humidity and ventilation rate and therefore not suitable for application in air quality control in livestock barns where relative humidity and ventilation rate vary widely in different seasons [2].

The counter electrode plate with a dimension of 30.5 cm × 5.1 cm × 0.6 cm (L × W × D) and 8 holes (2 cm in diameter) was made of polished aluminum sheet. The electrical

potential between the syringe needle and a counter-electrode plate was provided by a high-voltage power supply (Kasuga Denkie Inc., Model#: APM-30KIPNX, Japan).

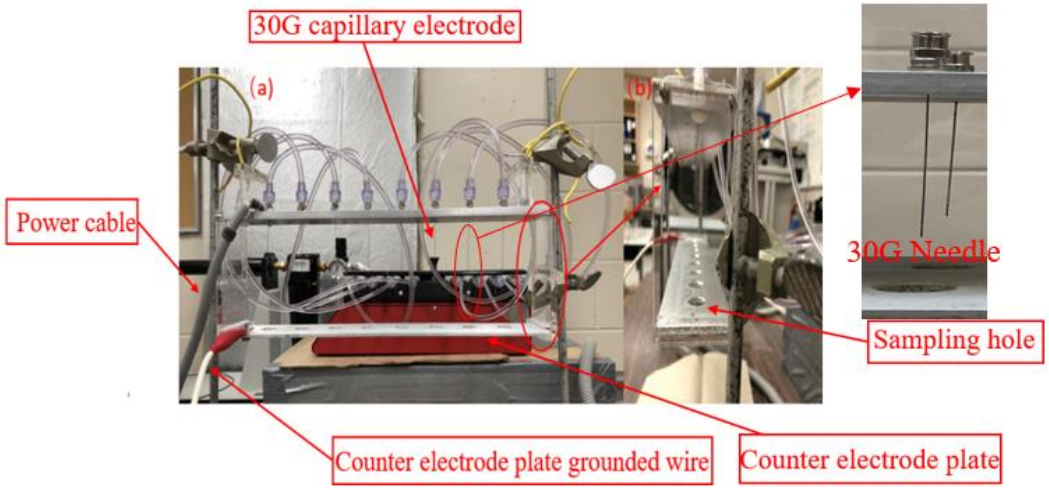

Figure 3.1 Picture of the prototype EWNS generator (a) Front view of generator with 8 units. (b) Side view of generator with slot for changing the electrode distance.

3.3.2 Size measurements of EWNS

Similar to previous studies [6, 68-69], Atomic Force Microscope (AFM), (4500 AFM, Keysight Technologies, USA) and Aspire CT170 probe (Nanoscience Instruments, Canada) were used to measure the size distribution of generated EWNS to ensure the droplets were in the nanoscale range at different operating conditions. The AFM scan rate was 1 Hz and the scanned area $5\mu m \times 5\mu m$ with 512 scan lines. All images were processed by the Gwyddion. The mica surface was placed at a 1.0 cm distance from the counter electrode for an average exposure duration of 15 seconds to avoid surface flooding caused by the droplets coalescence. The fresh cleaved mica surface was scanned immediately after the spray. The contact angle of water with a freshly cleaved mica surface is 0° [6, 68-69] The water droplets with dome-like shape were measured by AFM to acquire the physical dimension of the spread droplet [6, 68-69]. The method was used to calculate the final size of spread water droplet [68-69]. The equivalent diameter was calculated with the following equation:

$$\boldsymbol{d = \sqrt[3]{2h^2(3\frac{a^2+h^2}{2h} - h)}} \quad (3.1)$$

where *h* is the measured height and *a* is the measured radius of the spread droplet. In total 20 droplets were measured to determine the average size of EWNS [68], and the height and equivalent diameter of each individual droplet are shown in the supplementary material.

A range of operating parameters from Table 3.1 were tested at three different scenarios (i.e., changing electrode distance, flow rate, and pH) to explore the trends, which were used to confirm the mean EWNS droplet size at each tested condition was in the nanoscale (<500 nm) [71]. In Table 3.1, the range of electrode distance was from 2 cm to 4 cm, the range of flow rate was from 1 µl/min to 3 µl/min, and the range of pH was from 7 to 12. The applied voltage was adjusted to generate a stable electrospray at each electrospray condition. When one operational parameter was changed, the rest of the other parameters were kept the same.

Table 3.1 Tested electrospray conditions for EWNS size measurements

Electrospray Conditions	Electrospray Operational Parameters				Chemical Properties	
	V (kV)	L (cm)	D (cm)	Q (µl/min)	pH	Liquid
1	-7.5	4	1.3	1	7	RO
2	-6.6	3	1.3	1	7	RO
3	-5.5	2	1.3	1	7	RO
4	-5	2	1.3	3	7	RO
5	-6.6	2	1.3	2	7	RO
6	-5.5	2	1.3	1	7	RO
7	-3.6	0.5	1.3	0.9	7	RO
8	-3.6	0.5	1.3	0.9	9	NaOH/water
9	-3.6	0.5	1.3	0.9	12	NaOH/water

3.3.3 Determination of EWNS generation rate

The EWNS generation rate was measured by the condensation particle counter (*CPC*) (Model 3007, TSI, USA). The sampling flow rate was 100 cm^3/min, and the data log interval was 1 second. The EWNS generation rate of a single needle unit was measured by placing the end of the sampling tubing below the hole of counter electrode, the average generation rate was determined based on a total sampling time of one hour.

3.3.4 Swine and poultry barn dust reduction experiments

3.3.4.1 Dust samples preparation

Swine and poultry barn dust were selected as the test particles for the fine dust reduction in livestock buildings. Settled swine dust samples were collected from the top of the penning in six rooms at the swine barn facility of the Prairie Swine Centre (Saskatoon, Canada), the average age of pigs was 24-25 weeks and weight on average was 135 kg. Settled poultry dust samples were collected from the floor in different broiler rooms at the Poultry Centre (University of Saskatchewan, Saskatoon, Canada). Both swine and poultry barn dust samples were sieved into smaller size fraction by a 50 μm sieve (Canadian Standard Sieve Series, Ontario, Canada) before being loaded into the dust dispenser (RBG 1000 I, PALAS, Germany) to aerosolize the dust material inside the experimental chamber.

3.3.4.2 Experimental procedures

All experiments were conducted in an acrylic experimental chamber with dimensions of 1.0 m × 0.5 m × 0.5 m (L × W × D). Surfaces of both the experimental chamber and dust dispenser were grounded. Air exchange rates were precisely controlled by pressure gauges and mass flow meters. The air temperature and relative humidity (RH) were monitored throughout the experiments and the experiments were conducted at approximately 23°C and 45%, respectively. All tested conditions were conducted in triplicate and are represented by means and standard deviation.

Figure 3.2 shows the experimental setup. Part A is the dust generation system containing an air compressor (058-1292-2, Maximum, USA), a solid particle dispenser (RBG 1000 I, PALAS, Germany) supported with filtered dry and oil free air (Model 3074B, TSI, USA) at an inlet flow rate of 30 liters per minute (lpm) and 20 liters per minute (lpm) for a ventilation rate of 15 *ACH* with total air inlet flow rate of 62.5 lpm and 5 *ACH* with total air inlet flow rate of 20.8 lpm, respectively. The piston speed of the particle dispenser, which controls the dust feed rate, was adjusted to 12 mm/h (15 *ACH*) and 10 mm/h (5 *ACH*), respectively. Part B is the EWNS generation and dust reduction system, which consists of two EWNS generators (described in Section 3.3.1). Part C is the dust sampling and exhaust system, which includes an aerosol monitor (DusTrak DRX, TSI Inc) and a biosafety cabinet (B-2-4, Microzone, Canada).

For the control experiments, only the dust and HEPA-filtered air supply were injected into the experimental chamber. For the treatment experiments, the dust levels and air change rates were kept the same as control experiments, but with EWNS at varying operating parameters. Both control and treatment experiments were comparatively studied under controlled laboratory conditions to optimize the operating conditions in the treatment groups (Table 3.2). Such as, varying the spray liquid flow rate (i.e., 1, 2, and 4 µl/min), polarity of charges, applied voltage (i.e., ± 5.5 and ± 6.5 kV), electrodes distance (i.e., 2, 3, and 4 cm), and pH of spray liquid (i.e., 7, 9, and 12). The impact of EWNS on swine barn dust reduction was tested at two different air exchange rates (5 and 15 *ACH*), which are the common air change rates in the laboratory scale animal facilities [72]. This optimized condition was then tested at different pH for poultry barn dust reduction at 15 *ACH*. The total experimental time for both controls and treatments was 1 hour. For the control experiments, the average mass concentration of swine barn dust was maintained at 14.3 mg/m ^{3} at 5 *ACH*, and 12.5 mg/m ^{3} at 15 ACH. The average mass concentration of poultry barn dust was kept at 13.5 mg/m ^{3} at 15

ACH. It is worth mentioning that the average concentrations of control experiments were greater than reported total dust concentrations (~3.62 mg/m ^{3}) in swine confinement buildings and occupational exposure limit of 10 mg/m ^{3} of total dust [73, 74], which makes this study significant in terms of the impact of EWNS on fine dust reduction at levels higher than the occupational exposure limit.

Table 3.2 Tested operating conditions for swine barn dust reduction.

Operating Conditions	Electrospray Operating Parameters				Chemical Properties		
	V (kV)	L (cm)	D (cm)	Q (µl/min)	pH	Liquid	
1	5.5	2	1.3	2	7	RO	Changing voltage and polarity
2	6.5	2	1.3	2	7	RO	
3	-5.5	2	1.3	2	7	RO	
4	-6.5	2	1.3	2	7	RO	
5	6.5	2	1.3	1	7	RO	Changing flow rate
6	6.5	2	1.3	2	7	RO	
7	6.5	2	1.3	4	7	RO	
8	6.5	3	1.3	4	7	RO	Changing distance
9	6.5	2	1.3	4	7	RO	
10	6.5	4	1.3	4	7	RO	
11	6.5	2	1.3	4	7	RO	Changing pH
12	6.5	2	1.3	4	9	NaoH/water	
13	6.5	2	1.3	4	12	NaoH/water	

V: Applied voltage. *L*: Distance between capillary and counter electrode. *D*: sampling hole diameter on the counter electrode. *Q*: Flow rate of the sprayed liquids. RO: reverse osmosis water.

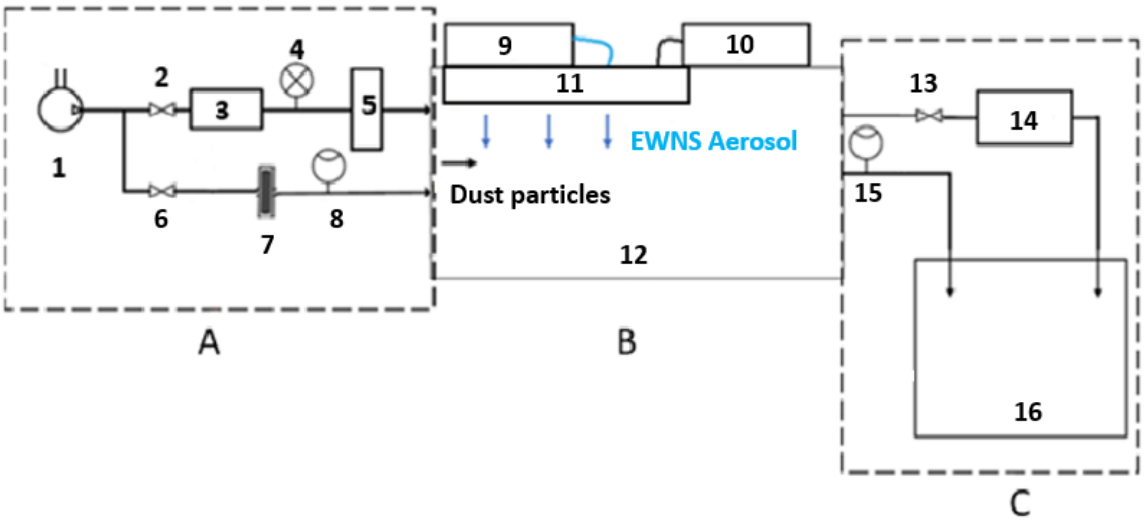

Figure 3.2 Experimental setup for fine dust reduction. A. Part 1 - dust generation system: 1. Air compressor; 2. Control valve; 3. Air filtration system; 4. Pressure gauge; 5. Dust dispenser; 6. Control valve; 7. HEPA filter; 8. Mass flow meter; B. Part 2 - EWNS generation and dust reduction system: 9. Syringe pumps; 10. Power supply; 11. EWNS generators: 12. Environmental chamber; C. Part 3 - dust sampling and exhaust system: 13. Control valve; 14. DusTrak aerosol monitor; 15. Mass flow meter; 16. Biosafety cabinet.

3.3.4.3 Airborne dust concentration analysis method

The airborne dust concentrations were measured by an aerosol monitor (DusTrak DRX, Model 8533, TSI, USA) with a measurable size range between 0.1 to 15 µm, sampling flow rate of 3.0 l/min and data log interval of 1 second and size range bands of PM1.0, PM1.0-2.5, PM2.5-4.0, PM4.0-10.0, and total dust (<15 µm). The effectiveness of the EWNS on the fine dust reduction was determined by comparing the dust concentration of control and treatment experiments. The dust reduction (η) was calculated using the following equation:

$$\eta = \frac{C_i - C_e}{C_i} \times 100 \quad (3.2)$$

where C_i is the average dust mass concentration of the control trials (mg/m ^{3}) and C_e is the average dust mass concentration of the treatment trials. A one-way ANOVA test was used to test for statistical differences between operating conditions with a level of significance of 5%.

3.4 Results and discussion

3.4.1 EWNS size

Table 3.3 shows the measured diameter of the droplets generated under each of varying operating conditions. The mean droplet size of all tested operating conditions was in the nanoscale range. In general, the mean EWNS droplet size follows the trends as described by the scaling law:

$$\boldsymbol{d_P \approx \left[\frac{(\beta-1)^{\frac{1}{2}} Q\, \varepsilon_0}{K}\right]^{1/3}} \tag{3.3}$$

where, d_P is the mean droplet size, β is relative permittivity, Q is liquid flow rate, ε_0 is vacuum permittivity, and K is electrical conductivity [75]. The smaller the flow rate, the smaller the droplets that were generated [75]. When the electrode distance was changed, the randomness of the droplet size distribution can be explained from the variation of the surface charge [76], the randomness of the Rayleigh effect and the inevitable evaporation over-time [6]. For the changing pH, the same trend was observed from a previous study [77] in which, as the liquid conductivity increased, the size of the EWNS produced by that solution increased.

Table 3.3 EWNS droplet size distribution at electrospray conditions described in Table 3.1

Electrospray Conditions	Droplet size (nm)
1	79±16
2	17 ± 4
3	45 ± 10
4	269 ± 30
5	263 ± 73
6	45 ± 10
7	16 ± 2
8	25 ± 7
9	73 ± 17

3.4.2 Total EWNS generation rate

The EWNS generation rate is a significant factor for the scaling-up of the process. Based on the condensation particle counter results, the EWNS generator, when run at condition 11 in Table 3.2, generated particles at a rate of 28,800 #/(cc min), and resulted in the highest total dust reduction (Table 3.4) with the most stable performance (smallest standard deviation). It is also noteworthy that the total water consumption of one EWNS generator at this optimal condition was 1.92 ml/h.

Table 3.4 Summary of the total swine barn dust reduction at all tested operating conditions from Table 3.2.

Operating Conditions	EWNS generation rate (#/(cc min))	Total dust reduction %
1	36480	66.6
2	24960	73.0
3	1836000	66.3
4	1051200	68.7
5	16800	61.9
6	29760	73.0
7	28800	73.0
8	11040	60.0
9	28800	73.0
10	17760	52.8
11	28800	73.0
12	24960	71.2
13	28800	72.3

3.4.3 Fine dust reduction performance

3.4.3.1 Verification of EWNS on swine barn fine dust reduction

Figure 3.3 shows the measured concentration of swine barn dust particles exiting the experimental chamber as a function of time as the prototype EWNS generator was turned on

and off at an air exchange rate of 15 *ACH*. Before the EWNS generator was turned on, the swine dust level increased and reached a steady state. After the EWNS generator was turned on, the dust level decreased dramatically and plateaued at 5 mg/m ^{3}. When the EWNS generator was turned off, the dust level increased back to the original level. Thus, it can be concluded that the EWNS contributed to the reduction of the swine barn dust.

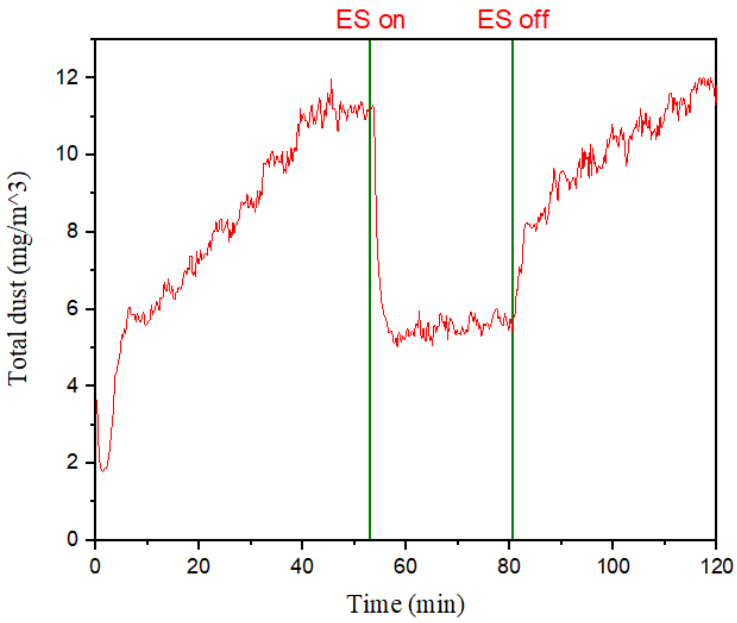

Figure 3.3 Measured swine barn fine dust concentration profile as affected by the operation of the EWNS generator. (ES on: the electrospray was on; ES off: the electrospray was off.)

3.4.3.2 Effect of applied voltage and polarity of charges

Figure 3.4 shows the highest total dust reduction and the most stable performance was observed at +6.5 kV. There was no statistically significant difference between the applied voltages in terms of total dust reduction ($p>0.05$) from non-parametric Kruskal Wallace test. However, +6.5 kV has a higher average total dust reduction and a smaller standard deviation than that for -6.5 kV (68.0% vs 73.0%). The results indicate that the polarity of charges carried by EWNS does not affect the performance of swine barn dust reduction at the uncharged dust contaminants condition (only EWNS were charged). By comparing with previous studies,

Almuhanna et al. [23] have demonstrate that negatively charged Wet Electrostatic Scrubber (WES) had higher removal efficiency than the positively charged WES for corn starch dust. Jaworek et al. [35] have shown that positively charged aerosols had higher suppression than negatively charged aerosols for cigarette smoke reduction. This is likely due to the difference of dust particle chemical compositions and size distributions that will affect the Coulombian attraction and contact efficiency between charged droplets and dust particles. Therefore, +6.5 kV was chosen as the optimal voltage for investigating the effects of other operating parameters.

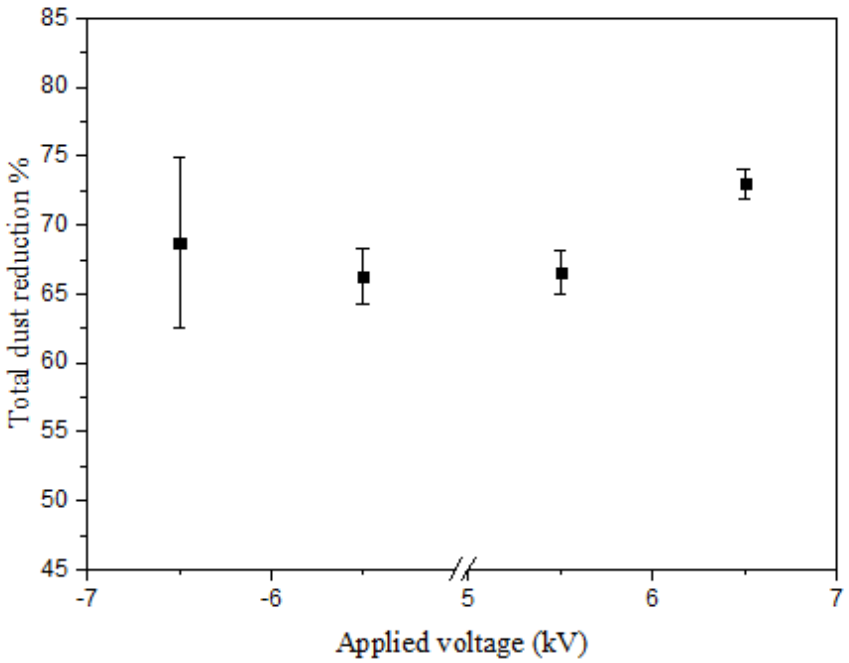

Figure 3.4 Total swine dust reduction at various applied voltage levels and air exchange rate of 15 *ACH* (Q = 2 µl/min, L = 2 cm, pH = 7, and spray time t = 1 hour).

3.4.3.3 Effect of sprayed liquid flow rate

Figure 3.5 shows similar total dust reduction was obtained for liquid flow rates of 2 µl/min and 4 µl/min (73.03% vs 72.90%, respectively). However, more stable performance (smaller fluctuation) was observed at 4 µl/min as compared to 2 µl/min. There is a significant difference of dust reduction between 1 and 2 µl/min as well as 1 and 4 µl/min ($p<0.05$) liquid flow rates. This is likely due to the differences in generated EWNS droplet size as presented in

Table 3.1, which showed that a much smaller mean droplet size (45 ± 10 nm) was generated from 1 µl/min compared to 2 and 4 µl/min (263 ± 73 and 269 ± 30 nm, respectively). Evaporation may also play a role as evaporation rate of EWNS droplets with mean size of 45 nm would be expected to be higher for this smaller droplet size than that for the larger EWNS droplets generated at 2 and 4 µl/min at 15 *ACH* of air exchange rate.

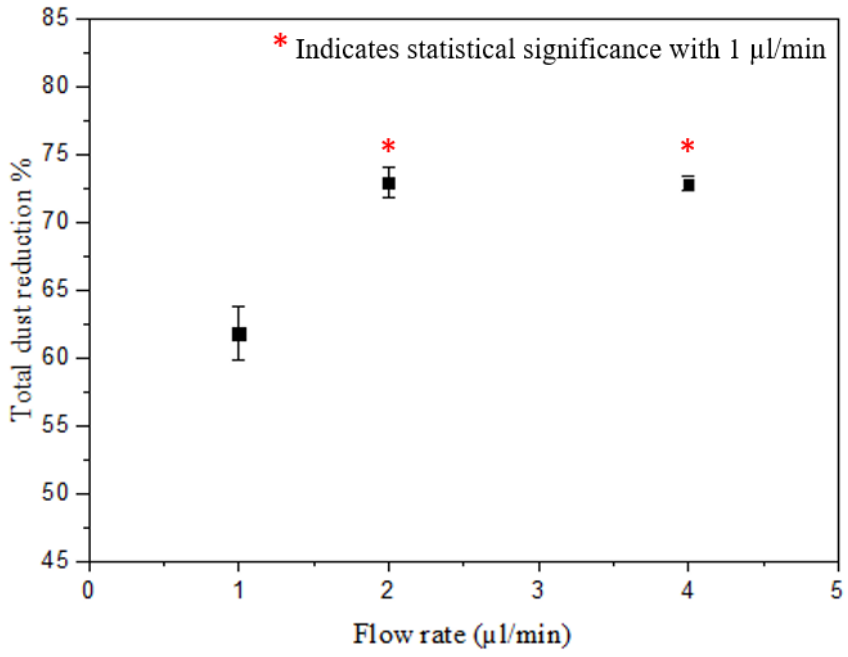

Figure 3.5 Total swine dust reduction at various spray liquid flow rates at an air exchange rate of 15 *ACH* (V = +6.5 kV, L = 2.0 cm, pH = 7, and t = 1 hour).

3.4.3.4 Effect of electrode distance

The effect of electrode distance on dust reduction is shown in Figure 3.6. It can be seen that the highest total dust reduction (75%) was achieved at a distance of 2.0 cm. There are statistically significant differences between the various electrode distances tested ($p<0.05$). This trend is consistent with previous fundamental study of electrospray that at constant voltage, a high electrical field can be generated through decreasing both capillary radius and electrode distance [78, 79]. When the distance is reduced, the generated electrical field will increase, resulting to increased fine dust removal as well [66, 80].

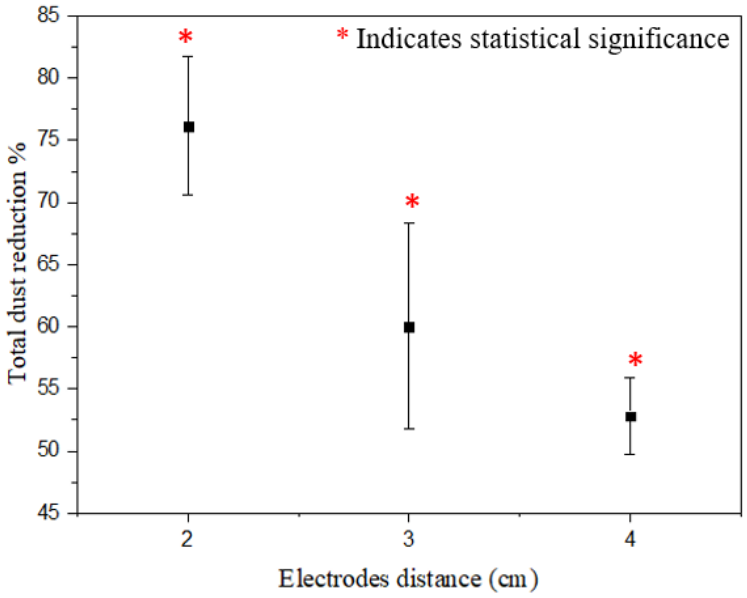

Figure 3.6 Total swine dust reduction at various electrode distances at an air exchange rate of 15 *ACH* (*V* = +6.5 kV, *Q* = 4 µl/min, pH = 7, and *t* = 1 hour). There is a significant difference between each tested electrode distance.

3.4.3.5 Effect of sprayed liquid pH

Figure 3.7 shows the effect of pH of spray liquid for the swine barn dust reduction. However, from the one-way ANOVA test, there was no significant difference between different pH levels ($p>0.05$). In order to explore the potential reason and find the optimum pH level for swine barn dust reduction, the reduction percent at different particle size bands were evaluated for both swine and poultry barn dust. Figure 3.8 shows results there is no clear trend between different pH at different particle size bands. However, for the poultry barn dust trials, it can be observed that the reduction increases as the pH increases at different size bands. From Table 3.4 and 3.5, the highest overall reduction (η=73%) and stable performance was observed at pH = 7 for the swine barn dust trials, whereas pH = 12 had the highest overall reduction (η=83%) for the poultry barn dust trials. The effect of pH on dust has been described by others including the relationship between sprayed liquid electrical conductivity and generated droplets current [9, 67]. From these studies, it was concluded that higher pH of spray liquid resulted in

higher electrical conductivity, which subsequently generated droplets with higher current and charge density. Droplets with higher charge density are beneficial for dust collection. However, the different behavior of the impact of pH on dust reduction observed in this study might be due to the differences of the chemical composition, surface structure, and charge distribution between swine and poultry barn dust particles that may affect the Coulombian attraction between EWNS and dust particles. Such as, the dust from broiler rooms has been shown to have lower concentrations of ash, potassium, chlorine and sodium than the pig rooms, while sodium and potassium concentrations were higher than pig rooms [81]. Surfaces of dust particles may also be important to the effects. Takai et al. have demonstrated that the particles from dairy houses have relatively smooth surfaces as compared to the particles from poultry houses, which have rough surfaces, and the structure of dust particles from farrowing house was between the two type of dust particles [82].

In order to verify the EWNS on fine dust removal at low ventilation rate, the optimized condition was tested for swine barn dust reduction at air exchange rate of 5 as well. Figures 3.10 and 3.11 show total dust concentration and reduction for different particle size bands. The experiments at 15 *ACH* show a slightly higher reduction than those observed at 5 *ACH* at each particle size band. This may be explained by the dust was removed by ventilation air more at higher ACH than lower *ACH*. However, there was no statistically significant difference between 5 and 15 *ACH*.

Table 3.5 Summary of the poultry barn total dust reduction at different pH.

Operating Conditions	EWNS generation rate (#/(cc min))	Total dust reduction %
pH 7	28,800	78.3
pH 9	24,960	80.6
pH 12	28,800	83.4

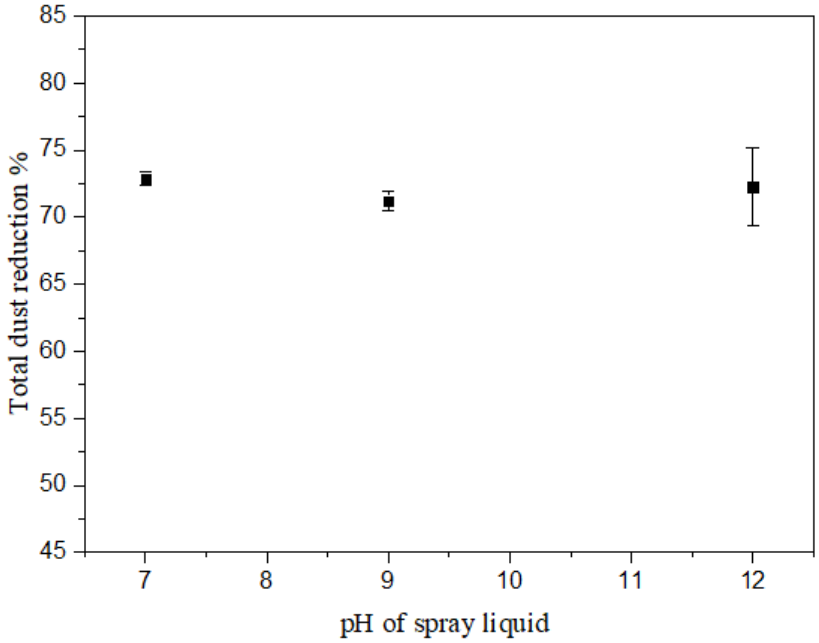

Figure 3.7 Total swine dust reduction at various spray liquid pH levels at an air exchange rate of 15 *ACH* (V = +6.5 kV, Q = 4 µl/min, L = 2 cm, and t = 1 hour)

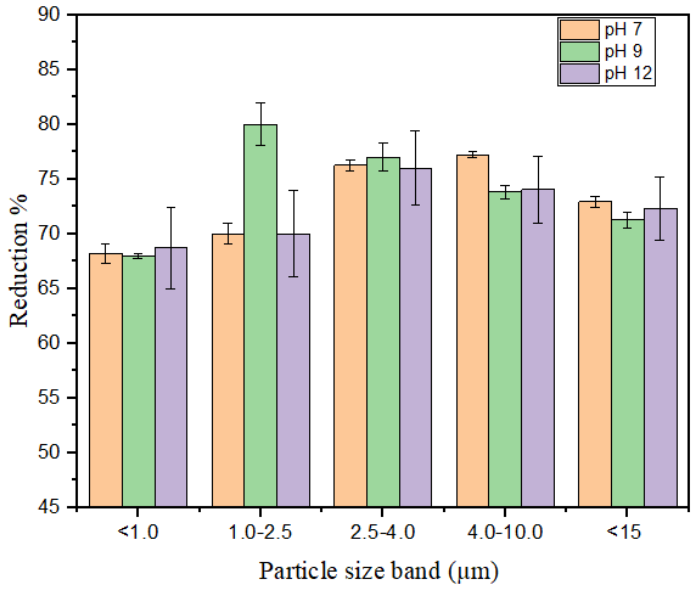

Figure 3.8 Reduction of the spray liquid with different pH for the swine barn dust at different particle size bands. The air exchange rate was 15 *ACH* (*V* = +6.5 kV, *Q* = 4 µl/min, *L* = 2 cm, and *t* = 1 hour).

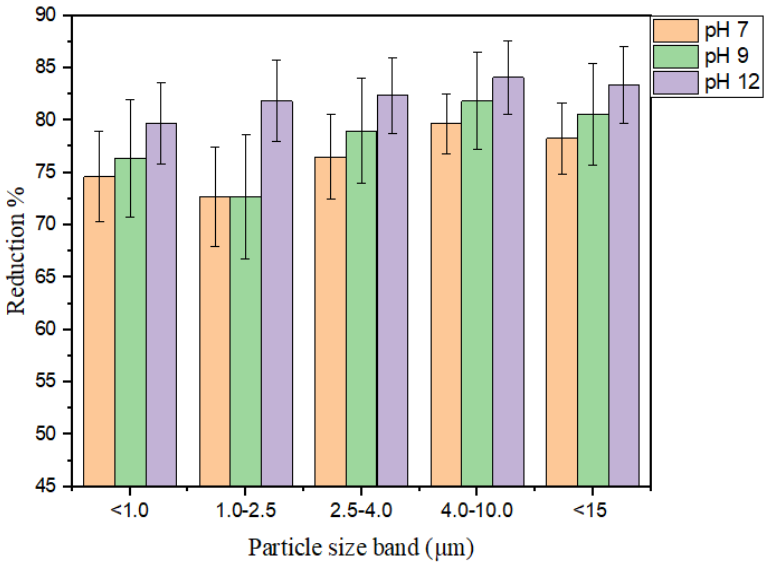

Figure 3.9 Reduction of the spray liquid with different pH levels for the poultry barn dust at different size bands. The air exchange rate was 15 *ACH* (*V* = +6.5 kV, *Q* = 4 µl/ min, *L* = 2 cm, and *t* = 1 hour).

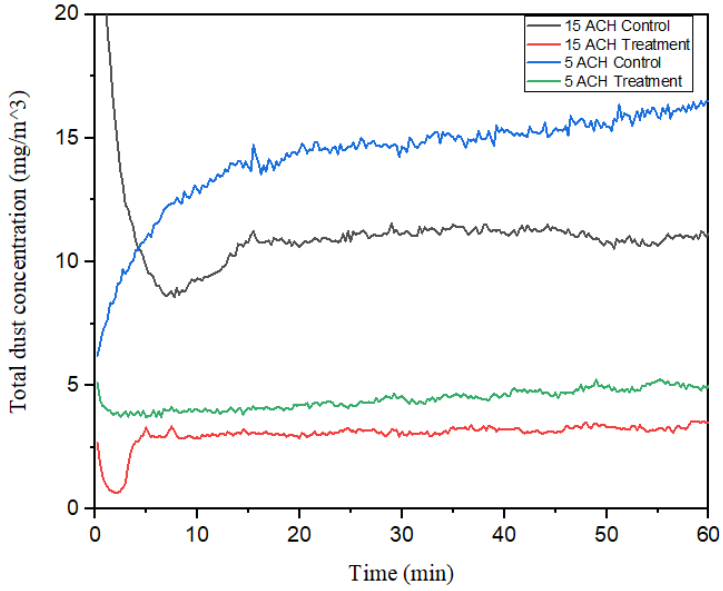

Figure 3.10 Average swine barn total dust mass concentration of control and treatment trials at air exchange rate of 5 and 15, respectively, V = +6.5 kV, Q = 4 µl/min, L = 2 cm, pH = 7, and t = 1 hour. Control: only the swine dust particles was injected, Treatment: both swine dust particles and EWNS aerosols were injected.

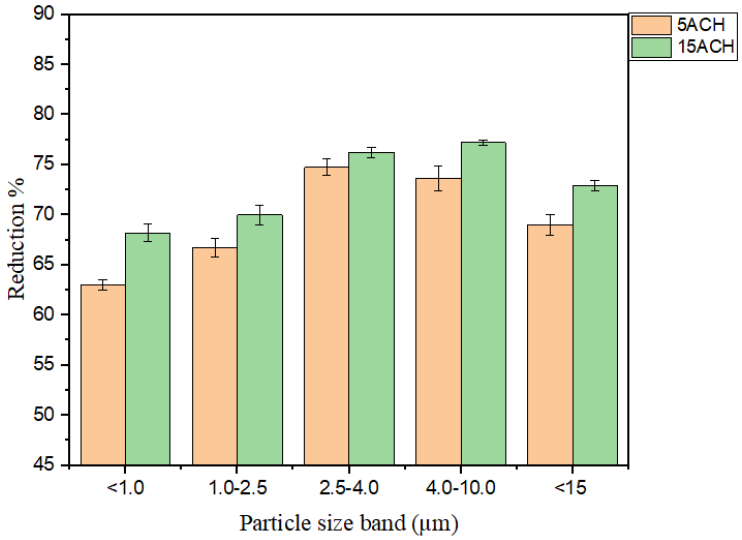

Figure 3.11 Swine barn dust reduction for different particle size bands at air exchange rate of 5 and 15, respectively, V = +6.5 kV, Q = 4 µl/min, L = 2 cm, pH = 7, and t = 1 hour.

3.5 Conclusions

A laboratory-scale dust removal technique using electro nano-spray (ENWS) was developed, tested and optimized for fine dust particles from swine and poultry barn operations. The following conclusions were drawn from this work:

- The fine dust reduction of EWNS was affected by the spray liquid flow rate, polarity of EWNS charges, and electrodes distance. The applied voltage did not have a significant impact on swine barn dust reduction, and the positively charged EWNS droplets had higher reduction than the negatively charged EWNS droplets for swine barn dust reduction. Moreover, there was no significant difference between two air exchange rates (5 and 15 air changes per hour) in terms of swine barn dust reduction.
- The removal was affected by the type of dust particle (i.e., composition, size distribution and charge distribution). The pH of spray liquid had a more profound impact on reduction of poultry barn dust than the swine barn dust.
- Under laboratory conditions, the total dust reduction of swine barn dust can reach up to 72.9% at the optimal operating condition of V = +6.5 kV, Q = 4 µl/min, L = 2 cm, and pH = 7. For the poultry barn dust, the highest reduction was 83.36% at the condition of V = +6.5 kV, Q = 4 µl/min, L = 2 cm and pH = 12.

3.6 Acknowledgments

Financial support from the Agriculture Development Fund (ADF) from the Ministry of Agriculture of Saskatchewan and Agrivita are greatly appreciated. RLee Prokopishyn is acknowledged for his support in the design and construction of EWNS generator.

Chapter 4 – Experimental Studies on Reduction of Bioaerosol inside Animal Confinement Buildings using Engineered Water Nanostructures Generated from an Electrospray

Contribution of the MSc student

The analytical approach was proposed by Eric Yang and Dr. Shelley Kirychuk and Thompson Brooke. Calculations and data analysis were performed by Eric Yang. Drs. Lifeng Zhang and Shelley Kirychuk supervised and provided consultation during the experiments and thesis preparation.

Contribution of this chapter to the overall study

In this chapter, the *E. coli* W3110 was selected as a representative strain to generate bioaerosol. The *E. coli* deactivation efficacy was examined under the most optimized operating condition from the previous study of using EWNS for surface deactivation. Three different sprayed liquids with different pH and electric conductivity were tested and the quantity of reactive oxygen species from different sprayed liquids were compared by using Electron Paramagnetic Resonance and water droplet size was characterized by using Atomic Force Microscopy. In addition, non-thermal plasma, which is another type of bacteria deactivation technology, was compared with the EWNS method on the *E. coli* deactivation.

4.1 Abstract

In this study, a newly designed prototype of an engineered water nanostructures generator was developed and tested under laboratory condition for bioaerosol inactivation in animal confinement buildings. Nano-scaled water droplets with size less than 100 nm were generated through the electro nano-spray. Reverse osmosis water was used as the spray liquid and *E. coli* W3110 was used as a representative strain for bioaerosol generation. This newly designed generator had a high EWNS generation concentration (up to 120,000 # cm^{-3}) with water consumption of only 480 µl/h to treat 29.2 and 62.5 liter per minute (lpm) of air flow rate. Three different sprayed liquids were employed to test the bioaerosol inactivation at two different air exchange rates, 7 and 15 air changes per hour (ACH). The experimental results revealed that sprayed liquid of pH 7 had the highest inactivation of 69% at 7 air changes per hour (ACH), and 37% at 15 air changes per hour (ACH). The experimental results show promise for electro nanospray as a means for bioaerosol inactivation in animal confinement buildings.

4.2 Introduction

Animal production systems have been increasingly changing with larger farms and greater number of animals produced per farm, with resultant increase in the production of indoor airborne contaminants and potential health risks [54]. The swine industry in Canada exported $465 million pigs in 2017 and the high demand in the market accelerated the industry movement to large-scale operations [83-84]. The increased number of pigs per building increases the level of air contaminants to which workers are exposed, and may result in adverse health effects for workers [85-86]. Chronic bronchitis, asthma, reduced lung function and infections have been shown in barn workers and associated with inhalation of workplace bioaerosols [87-89].

Reduction in bioaerosol loads is important to improving the air quality in livestock barns. Current air cleaning technologies employed in swine barns include the following: (1) ventilation is a common control, however, increasing ventilation to reduce bioaerosol concentrations is often not feasible due to temperature and climate considerations [19]; (2) spraying oil and soap mixtures into the air for short periods of time, several times a day. Oil spraying has been shown to be effective in reducing total dust levels, however, the process significantly increased the cleaning time between herds and exposures for workers [21-22]; (3) using a fiber-based air purification system. This technology is effective in lowering the amount of bioaerosols emitted outside the building, however, it has less effect on indoor air quality, and the filtering material requires regular cleaning and maintenance, which increases operational costs [19].

A novel, nanotechnology-based, air purifying technique using Engineered Water Nanostructures (EWNS) generated from electrospraying was tested as a potential method for bioaerosol reduction in swine confinement buildings in this work. Electrospraying is a widely used technology for aerosol generation with a controlled size range. The electrospraying process can be divided into two steps. For step 1, a high voltage was applied between a needle capillary and a counter electrode plate. The high electrical potential will cause either positive or negative charges accumulate at the tip of the needle capillary. When the electrostatic force overcome the force from surface tension of the spray liquid, the spray liquid then will be pulled toward the counter-electrode plate and a spray mode will start to form [8]. For step 2, due to the Rayleigh effect, droplets carry same type of charges will repel each other, and split into finer and finer droplets [9-10]. Operating parameter, such as applied voltage, spray liquid properties, the capillary diameter, and the spray liquid flow rate, play an essential role for the electrosprayed droplet size distribution [67]. Applications of the nanoscale water droplets have been recently demonstrated in various fields, such as airborne bacteria and virus inactivation

in residential buildings, improvement of anti-pilling property of knitted wool fabric, and hotpot odor removal [53, 68, 69, 90]. From a bioaerosol inactivation perspective, the EWNS possess some unique physicochemical properties [53, 77, 90-92]. These properties including highly charged and mobile droplets with longer lifetime, and as reactive oxygen species carriers will be beneficial for bioaerosol inactivation applications [40, 93]. More importantly, this is a green chemical process that leaves no chemical residues as well as a toxicologically benign method, which does not pose potential risks to surrounding environment and barn workers [53]. However, there are no studies on the effectiveness of bioaerosol inactivation under conditions related to livestock buildings, which have much larger air exchange rates, higher relative humidity, and higher bioaerosols concentration than residential buildings [2, 94-96].

The objectives of this study were to develop an electro nano-spray platform and to test inactivation of bioaerosol under controlled conditions relevant to livestock operations.

4.3 Materials and methods

4.3.1 EWNS generator

For the EWNS generator, the same prototype of EWNS generator for the swine and poultry dust reduction was also used for airborne *E. coli* inactivation in this chapter. For the details of the EWNS design can refer to Chapter 3.3.1 in this thesis.

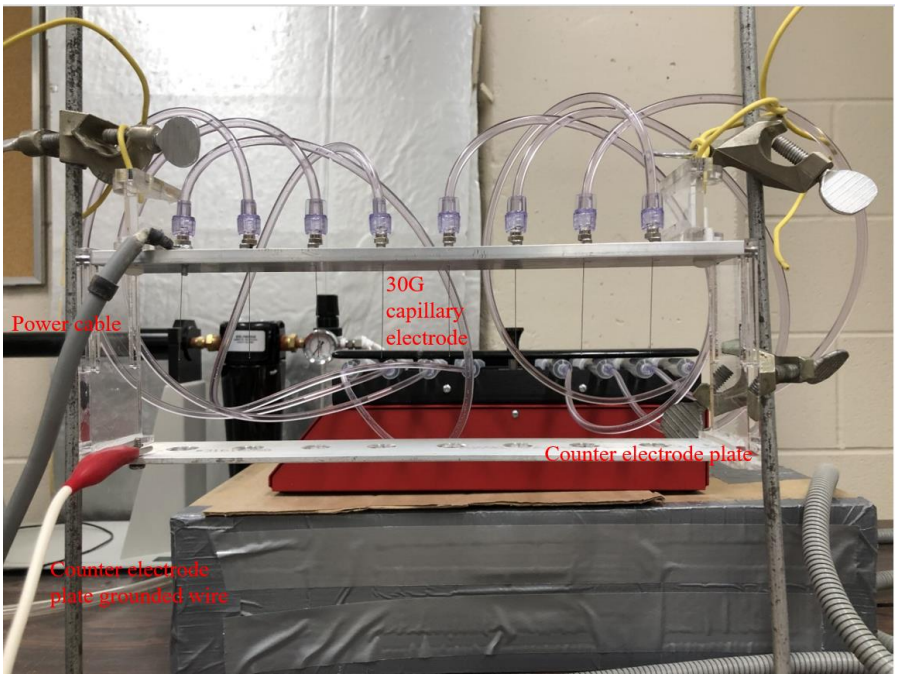

Figure 4.1 Picture of the prototype EWNS generator.

4.3.2 EWNS physicochemical characterization

4..3.2.1 Size measurements of EWNS

For the size measurements of EWNS, the same experimental method and instrument as the Chapter 3 were used. For details, it can be referred to Chapter 3.3.2 in this thesis.

4.3.2.2 Lifetime measurements of airborne EWNS

In order to investigate the lifetime of the airborne EWNS, both the inlet and outlet of the experimental chamber were closed. The EWNS generator was turned on for 30 minutes to continuously produce EWNS aerosol inside the chamber. After 30 minutes, the EWNS generator was turned off, and the airborne EWNS aerosol concentration was measured by a condensation particle counter (CPC) (Model 3007, TSI, USA). The sampling flow rate was 100 cm^3/min, and the data log interval was 1 second. The total measuring time was 5 hours. The chamber walls were grounded to minimize electrostatic particle losses.

4.3.2.3 ROS characterization of EWNS

Electron Spin Resonance (ESR) spin trapping was applied to detect the presence of short-lived free radical intermediates that include hydroxyl and superoxide radicals in the EWNS [68, 69, 97]. The 5,5-dimethyl-1-pryroline-N-oxide (DMPO), a widely used spin-trapping agent was used for the detection of hydroxyl and superoxide radicals [68, 98-99]. DMPO was obtained from Enzo Life Sciences (FroggaBio Scientific Solutions, Ontario, Canada). The Bruker EMX spectrometer (Bruker Instruments Inc, Billerica, MA, USA) was used for the detection of radicals. For detection of OH· and $O_2^{\cdot-}$ radicals, 30 µl of DMPO was dissolved in 1 ml Millipore water and ethanol, respectively. The DMPO solutions were poured into 2 ml centrifuge tubes and placed below the EWNS generator for a period of 20 minutes. After 20 minutes of sampling, 50 µl of trapping solution was transferred into a melting tube (Thomas Scientific, Swedesboro, USA) and placed into an EPR sample tube (Thomas Scientific, Swedesboro, USA) for detection. The data acquisition program Xenon 1.1b50 (Bruker Instruments Inc, Billerica, MA, USA) was used for data analysis.

4.3.3 Bioaerosol inactivation experiments

4.3.3.1 Bioaerosol type and generation system

Escherichia coli was selected as a representative test bioaerosol since it is a common pathogen found in swine confinement buildings [95]. Furthermore, it is widely accepted as a test aerosol and used to evaluate the bacteria inactivation for various techniques, such as thermal energy and photocatalytic inactivation [100-101].

For the bioaerosol generation, a CN-6 Collison nebulizer (BGI Inc., Pacwill Environmental, Canada) operated with HEPA-filtered dry and oil free air at absolute pressure of 20 psi (138 kPa) was used to aerosolize an *E. coli* solution. The concentration of the stock solution was optimized to get the optical density of 1, followed by a five-hundred-fold dilution using autoclaved deionized water to produce airborne bacteria concentrations of $2.0 \times 10^4 \pm 5$

777 CFU/m^3 (colony forming units per cubic meter) at an air exchange rate of 7 air changes per hour (ACH), which is close to the reported airborne *E. coli* concentration found in swine confinement buildings [95].

4.3.3.2 Bioaerosol inactivation experiments protocol

For the experimental setup, a similar experimental design as the one in chapter 3 was used and it can refer to Figure 3.2, two modules of EWNS generators were attached at the top of a 250 L acrylic chamber, the flow rate of each electrospray unit was precisely controlled by an 8-channel multi-syringe pump (NE-1800, NEWERA, USA). Each unit was equipped with a 30G needle (Needle Metal Hub, Hamilton, USA). The electrical potential was provided by a high-voltage power supply (Kasuga Denkie Inc., Model#: APM-30KIPNX, Japan). All HEPA-filtered air streams were supplied by an air compressor (058-1292-2, Maximum, USA). The bioaerosol with the mass flow rate of 12 litres per minute (lpm) was injected from the left side of the chamber, the two aerosol streams were mixed in the chamber and the number of culturable airborne bacteria was monitored as a function of time. Three different sprayed liquids (see Table 4.1) were tested at 7 ACH, and the sprayed liquid with the highest inactivation and the most stable performance was tested at a higher air change rate of 15 *ACH*. Both 7 and 15 air changes per hour (*ACH*) are the common ventilation rates used in prior laboratory scale animal facilities experiments [72]. During all experiments, the temperature and relative humidity in the chamber were monitored by a temperature and relative humidity probe (HIH8120-021-001, Humidlcon, Honeywell, USA), and maintained at 20-25 °*C* and 40-50%, respectively. The ozone concentration inside the chamber was measured by ozone detector tubes (GAS18L, Gastec, Japan) attached to a gas sampling pump (GV110, Gastec, Japan).

In summary, the protocol for the experiments was as follows: the two aerosols started at t = 0 min. The aerosol and main air stream inlets were controlled to have the total flow rate

reach 29.17 lpm (7 air changes per hour) and 62.50 lpm (15 air changes per hour). An air sample was obtained using a six-stage ambient viable impactor (TE-10-830, Pacwill Environmental, Canada) at three time-intervals during the total treatment time of one hour. All air samples were collected on lysogeny broth agar (LBA) plates, at a sampling flow rate of 28.3 lpm with a sampling time of 1 minute. Prior to collection of air samples, all stages of the viable impactor were sanitized by 70% isopropyl alcohol to eliminate any potential microbial cross-contamination. Post sampling the culture plates were incubated for 48 h at ambient conditions prior to colony counting. Colony counting was corrected by the positive hole corrections [102].

The total airborne *E. coli* concentration was calculated as the sum of the *E. coli* concentration of all six size intervals in a six-stage ambient viable impactor, using Equation (4.2). The total *E. coli* inactivation percent was calculated using Equation (4.3).

$$C_{TB} = \sum_{i=1}^{6} C_{Bi} \tag{4.2}$$

Where C_{TB} is the total airborne bacteria concentration (CFU/m^3) and C_{Bi} is the concentration of airborne bacteria in size interval *i* (CFU/m^3).

$$\text{Total inactivation percent} = \frac{C_{TBC} - C_{TBT}}{C_{TBC}} \times 100 \tag{4.3}$$

Where C_{TBC} is the total airborne bacteria concentration (CFU/m^3) in control trial and C_{TBT} is the total airborne bacteria concentration (CFU/m^3) in EWNS treatment trial.

For the control experiments, only the bioaerosol and HEPA-filtered air were injected into the experimental chamber and the EWNS generators inside chamber were turned off., with all other procedures, sampling and culturing protocols being kept the same.

Table 4.1 The operational parameters and main physicochemical properties of EWNS.

Electrospray Condition	Chemical Properties		Physical Properties
	pH	Liquid	Diameter (nm)
1[a]	7	RO	45 ± 10
2[a]	12	NaOH/water	40 ± 5
3[a]	7	0.9% saline	N/A

V: Applied voltage. *L*: Distance between electrode and counter electrode. *D*: Diameter of counter electrode hole. *Q*: Flow rate of sprayed liquid. *RO*: Reverse osmosis water. N/A: Not available. a: V = -5.5 kV, L = 2 cm, D = 1.3 cm, Q = 1 µl/min.

4.4 Results and discussion

4.4.1 Bioaerosol size distribution

Characterization of airborne *E. coli* particle size distribution is important as the submicron particles, such as particles smaller than 5 µm penetrate into the lower respiratory track and lungs that is important to health outcomes experienced by exposed livestock workers [103]. Thus, in this study, the aerosolized bacteria with sizes smaller than 10 µm was desired. Figure 4.2 shows 90% of the viable bioaerosol was recovered on Stage #5 (2.1-1.1 µm) and Stage #6 (1.1-0.65 µm) of the six-stage viable impactor. This result is similar to the pervious study on aerosolized bacteria from different suspending media, which demonstrate nearly all the airborne bacteria suspended in water-only medium were collected on Stage #6 of an ambient viable impactor [24].

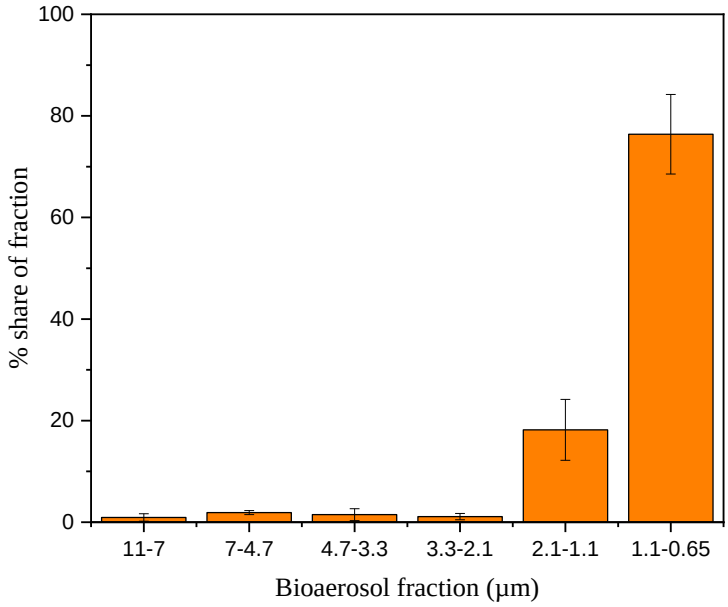

Figure 4.2 The aerosolized E. coli size distribution with suspension medium of deionized water

4.4.2 EWNS size measurements

Figure 4.3 shows the AFM topographies of conditions 1 and 2 (Table 4.1) on the mica surface with a 5 µm × 5 µm scanning area after EWNS spray, the equivalent average diameter of EWNS of conditions 1 and 2 were estimated to be 45 ± 10 and 40 ± 5 nm, respectively (original data can be found in the Supplementary Material). From Figure 4.4, it can be concluded that the droplet size of condition 3 can not be characterized, because the spread water droplets have NaCl salts (indicated by the red arrow in Figure 4.4), which were scanned by the AFM as well. Thus, the saline water droplets can not be distinguished from these NaCl salts.

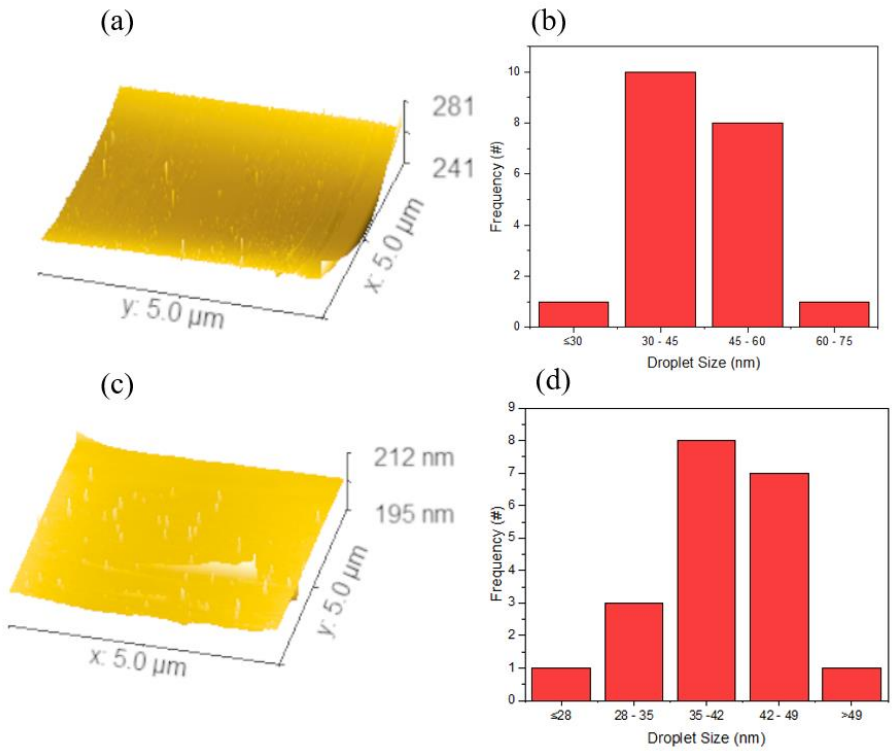

Figure 4.3 AFM measured size distribution of condition 1 **(a)** A 3D AFM image of RO water droplets as spread on a mica surface. **(b)** The calculated size distribution of the RO water droplets based on AFM measurements. **(c)** A 3D AFM image of NaOH/water droplets as spread on a mica surface. **(d)** The calculated size distribution of the NaOH/water droplets based on AFM measurements.

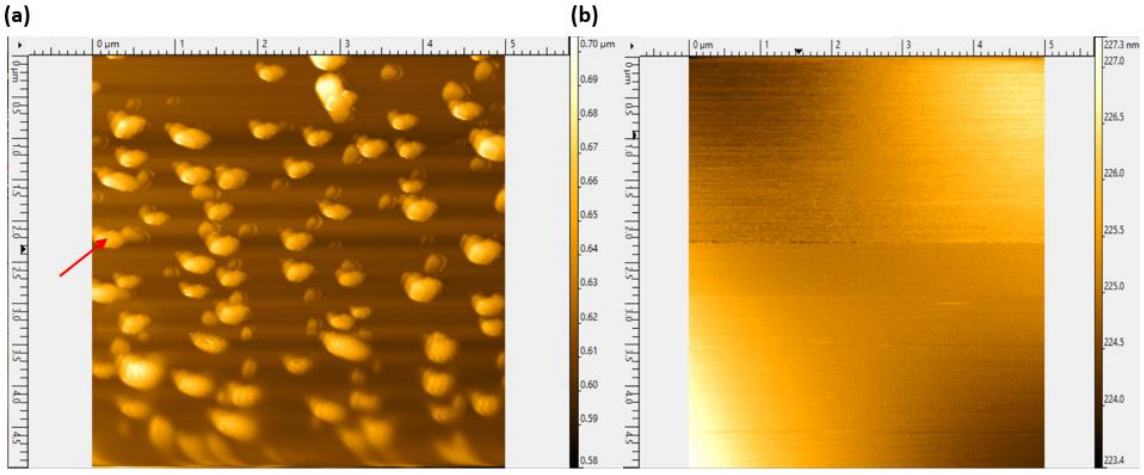

Figure 4.4 AFM measured size distribution of condition 3 **(a)** A 2D AFM image of saline water droplet as spread on a mica surface. **(b)** A 2D AFM image of fresh cleaved mica surface.

4.4.3 EWNS lifetime measurements

Figure 4.5 shows the concentration of airborne EWNS from different spray liquids in a closed 250 L chamber as a function of time. After the EWNS generator was turned off, the EWNS aerosol concentration still continued to increase for 10 to 30 minutes. This phenomenon can be explained by the Rayleigh effect, wherein the droplets with high charge density kept reducing its size until the charge reached equilibrium state [104]. Moreover, the spray liquid consumption of each EWNS generator was only 480 µl/min, therefore, there is no potential risk of barn floor flooding and no post-treatment of the aqueous solution is required.

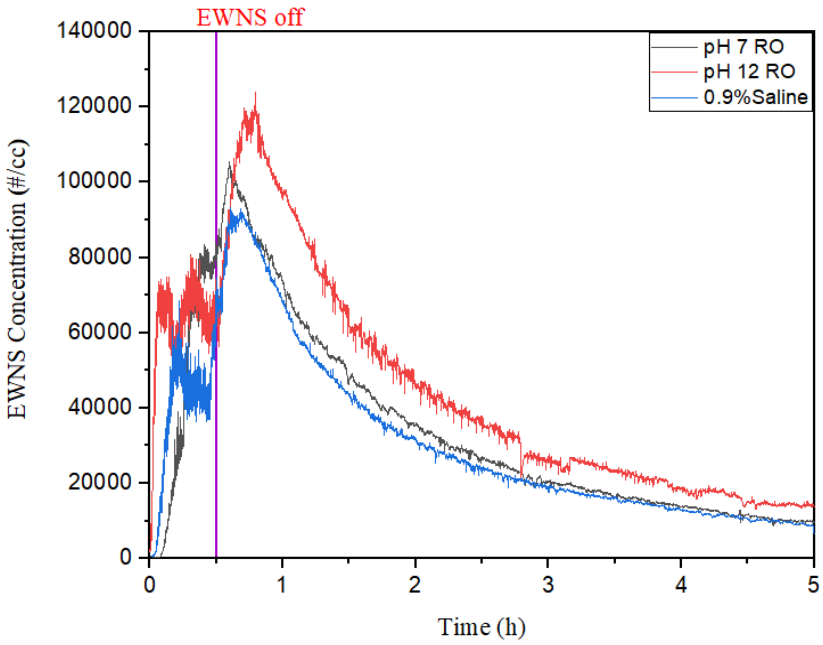

Figure 4.5 Concentration of airborne EWNS from different spray liquids as a function of time.

As shown in Figure 4.5, the pH 12 RO water had the highest EWNS concentration compared to pH 7 RO water and 0.9% saline water. Over the lifetime of EWNS, even after 4.5 hours, there were still EWNS droplets present in the chamber for all sprayed liquid conditions. The airborne EWNS degradation rates were found to be 18,604, 20,930, and 25,000 #/cc·h for 0.9% saline, pH 7 RO, and pH 12 RO, respectively, which were not significantly different among the three sprayed liquids.

4.4.4 ROS characterization of EWNS

The ESR spectra of OH· for different treatments (Table 4.1) are presented in Figure 4.6. The ESR spectrum clearly indicates the presence of OH· in EWNS, which has been reported in previous studies [68-69]. The signal intensity of OH· from pH 12 RO water was the highest. There was no clear difference OH· intensity between pH 7 RO water and 0.9% saline. The literature values were used to simulated ESR spectra of $O_2^{\cdot-}$ [97]. The simulated and measured ESR spectra of $O_2^{\cdot-}$ at the same treatments are presented in Figures 4.7 and 4.8; comparison of the simulated and measured spectra showed that the signal intensity of $O_2^{\cdot-}$ was very weak (almost the same level as background noise) for all treatment conditions. This indicates that OH· is the predominant radical species while $O_2^{\cdot-}$ is present in a smaller amount. Similar findings were reported by others [6]. Moreover, due to the rapid decay of DMPO./OOH into DMPO./OH, the $O_2^{\cdot-}$ that exists in the solution will convert quickly into OH· [97, 99].

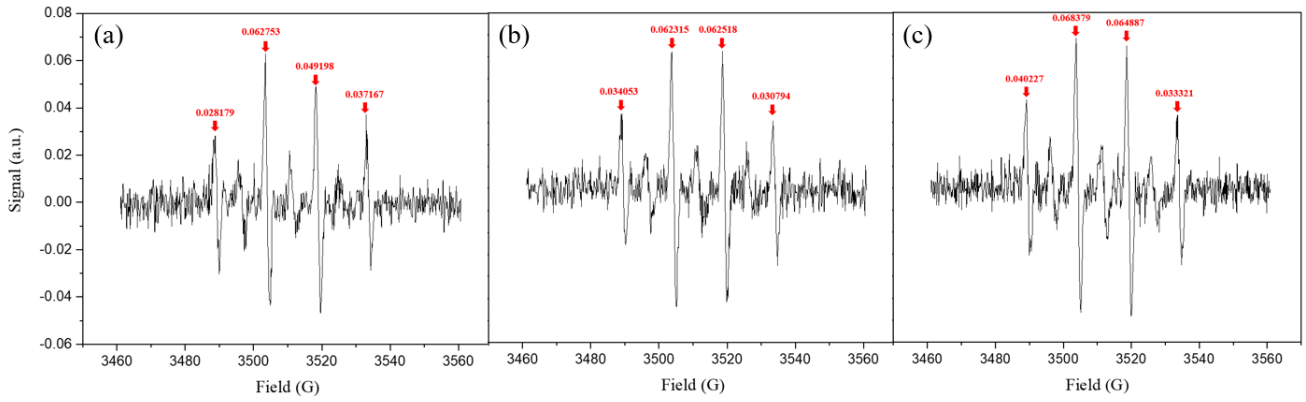

Figure 4.6 OH· characterization of EWNS. (a) EWNS treatment of using pH 7 RO water. (b) EWNS treatment of 0.9% saline. (c) EWNS treatment of using pH 12 RO water.

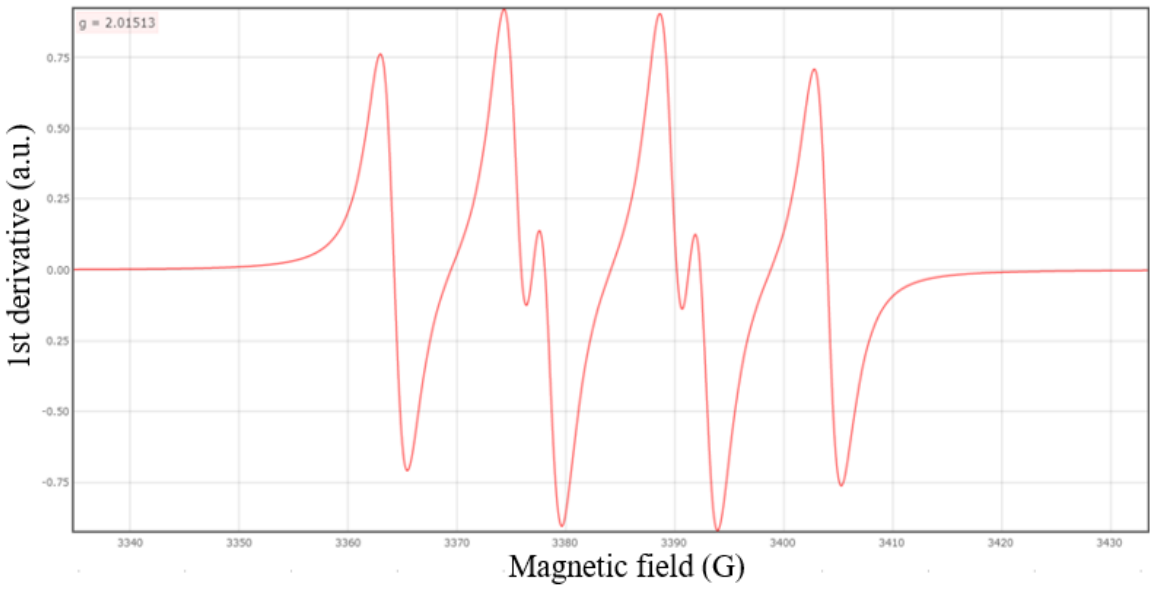

Figure 4.7 Simulated ESR spectrum of $O_2^{\cdot-}$.

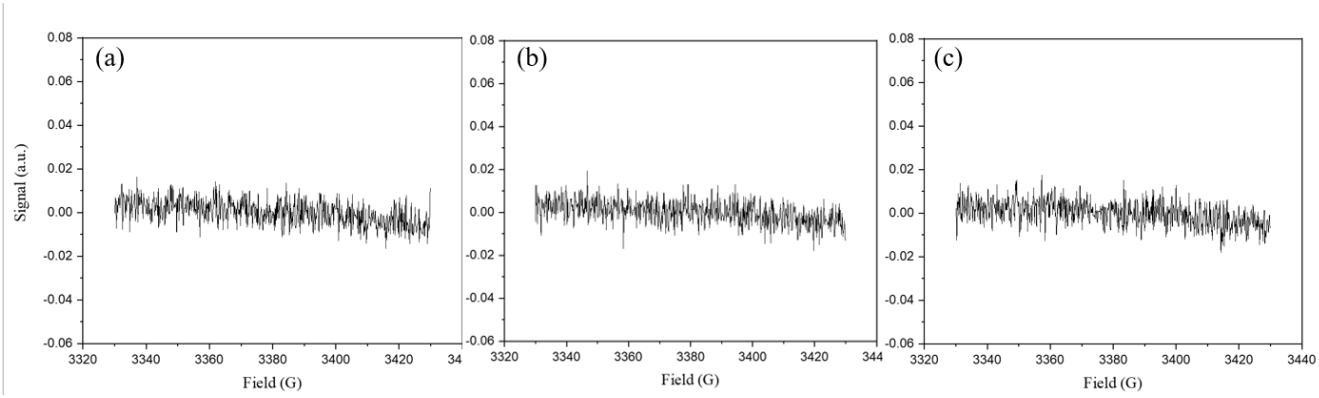

Figure 4.8 $O_2^{\cdot-}$ characterization of EWNS. (a) EWNS treatment of using pH 7 RO water. (b) EWNS treatment of 0.9% saline. (c) EWNS treatment of using pH 12 RO water.

4.4.5 Bioaerosol inactivation experiments

In order to ensure only the EWNS contributed to the airborne *E. coli* inactivation, a verification experiment was done. Figure 4.9 shows the viable concentration collected at the exit of the chamber as a function of time when the electro nano-spray was turned on at time t = 60 minutes and turned off at t = 100 minutes, under an air exchange rate of 7 *ACH*. The bioaerosol concentration decreased when EWNS was turned on. Once the EWNS was turned off, the bioaerosol concentration started to increase again. Because of the long lifetime of

EWNS, there were still residual EWNS droplets suspended in the chamber after the EWNS was turned off, thus longer time was needed for the bioaerosol concentration to increase back to the original level.

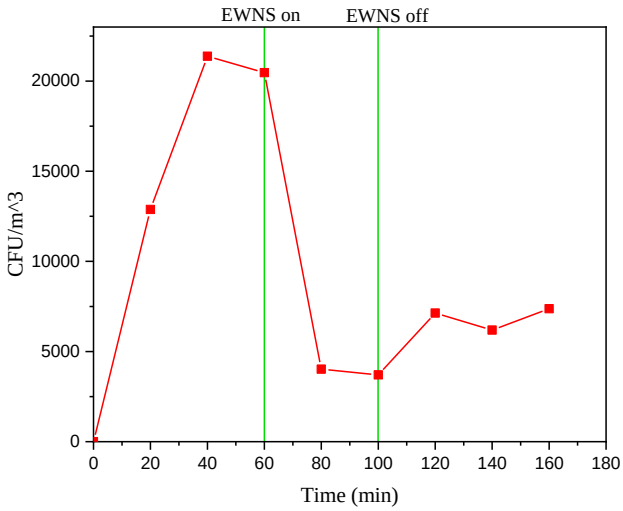

Figure 4.9 The bioaerosol concentration profile influenced by EWNS. (EWNS on: the electrospray was on; EWNS off: the electrospray was off.)

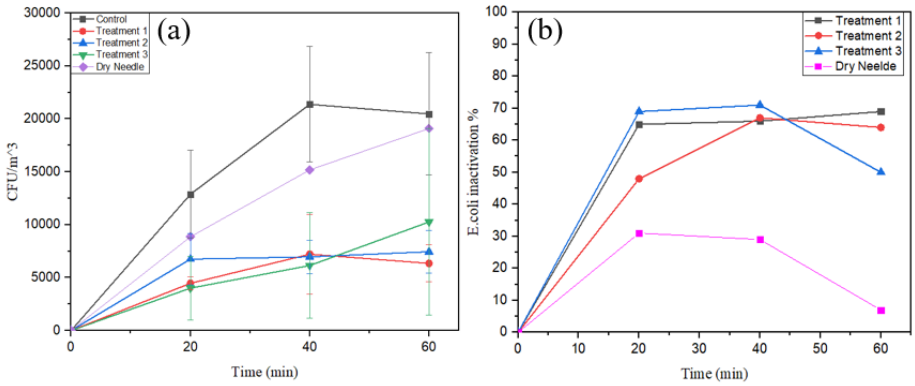

Figure 4.10 Bioaerosol inactivation reuslts. (a) The E. coli inactivation experiments at 7 air changes per hour (*ACH*). (b) The E. coli inactivation efficency results at 7 air changes per hour (*ACH*). Treatment 1: pH 7 reverse osmosis water. Treatment 2: 0.9% saline water. Treatment 3: pH 12 reverse msmosis water. Dry Needle: no spray liquid, only 1 µl/min of air was fed into the capillary.

Figure 4.10 presents the inactivation results of three different treatments at the ventilation rate of 7 *ACH*. After 1 hour, the EWNS reduced the culturable *E. coli* by 69% (pH 7 RO water), 64% (0.9% Saline), 50% (pH 12 RO water) and 7% (Dry Needle) compared to the control experiments. Before the steady state (40 min) was reached, the EWNS with the sprayed liquid of pH 12 RO water showed the highest *E. coli* inactivation. This can be explained by the relatively higher concentration of reactive oxygen species (ROS) in treatment 3 (pH 12 EWNS) than treatments 1 and 2. However, in terms of stability of inactivation, treatment 3 was decreasing compared with treatment 1 and 2. This may be explained by the differences in droplet size and charge density of EWNS between treatment 1 and 3 that affected contact efficiency (contact surface area, contact time) with bioaerosol droplets. Meanwhile, it can be noted that the Dry needle condition (equivalent to cold plasma) also contributed to the total *E. coli* inactivation. However, when the EWNS was applied at the same time, the inactivation was higher, generally twice of that observed under the dry needle condition.

The pH 7 RO water with the highest bioaerosol inactivation at 7 *ACH* was tested at a higher ventilation rate of 15 *ACH* as well. Figure 4.11 shows the airborne *E. coli* concentration at the control and treatment conditions at two different ventilation rates (7 and 15 *ACH*). The *E. coli* inactivation of treatment 1 at 15 air changes per hour (*ACH*) was only of 37% compared with 69% at 7 air changes per hour (*ACH*). This result is consistent with the previous study of EWNS inactivation on *Serratia marcescens* for residential air quality control, which found the inactivation of EWNS on bioaerosol inactivation depends on the residence time and evaporation rate of the EWNS inside the chamber that is mainly affected by the air exchange rate (*ACH*) [6]. The higher *ACH* used, the shorter residence time and the higher evaporation rate, resulting in lower bioaerosol inactivation.

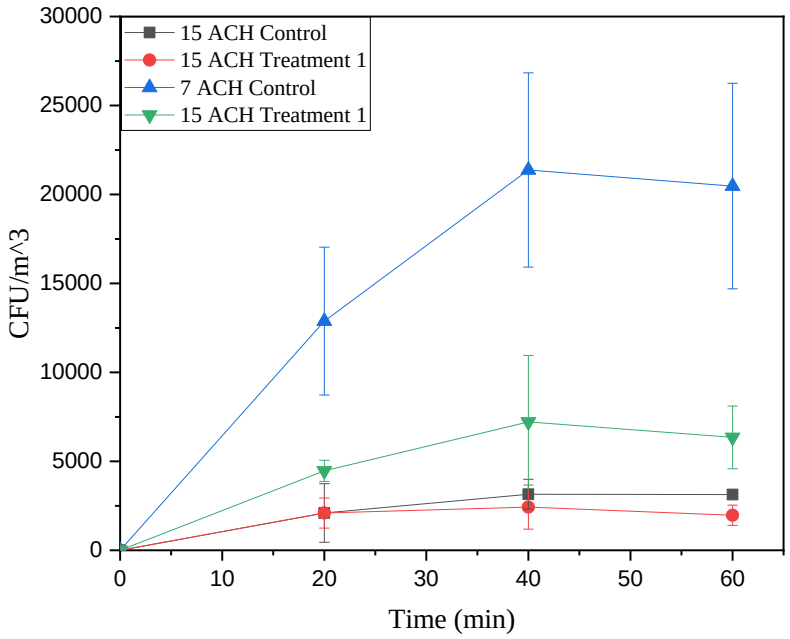

Figure 4.11 E. coli inactivation result of Treatment 1 (pH 7 reverse osmosis water) at 7 and 15 air changes per hour (*ACH*), respectively.

Table 4.2 The E. coli inactivation efficiencies of different EWNS treatments at 7 air changes per hour.

Size fraction (μm)	Bioaerosol inactivation (%)		
	pH 7 RO	pH 12 RO	0.9% Saline
11-7	-29±81	-53±84	-267±218
7-4.7	85±19	42±71	-12±66
4.7-3.3	41±35	9±21	-32±58
3.3-2.1	6±72	6±58	-42±92
2.1-1.1	67±16	70±9	30±35
1.1-0.65	72±8	66±9	43±39

As shown in Table 4.2, it is seen that the pH 7 RO water showed the highest inactivation and the most stable performance at each bioaerosol size fraction. Moreover, the nano-scale EWNS droplets (see the data in Table 1) tend to have better inactivation efficacy at smaller

size fractions (2.1-1.1 μm and 1.1-0.65 μm) when compared to a larger size fraction (11-2.1 μm), which had a large variation of reduction for all EWNS treatments. This may be explained by the lesser number of colony forming units on stages 1 to 4 relative to stages 5 to 6, such that a small variation in the number of colony forming units can change the calculated reduction percent considerably. Moreover, due to the long spray time, coagulation of particles might happen, which affected the inactivation for larger size fractions more [105]. During the experiments, the ozone level in the chamber for all EWNS inactivation experiments was monitored and generally the concentration was lower than 50 ppb (the lowest detection limit of ozone tube), which is not high enough to cause any bacteria inactivation [106]. Therefore, this confirms that only EWNS contributed to the bioaerosol inactivation.

4.5 Conclusions

The present study showed that the performance of the newly designed EWNS generator was stable and the spray liquid consumption was only 960 μl/min (Two EWNS generators) to treat 29.2 and 62.5 liter per minute (lpm) of air flow rate. The EWNS produced by this EWNS generator is effective in deactivating bioaerosol at high concentrations of up to 2.5×10^4 CFU/m^3, achieving *E. coli* inactivation up to 69% at 7 air changes per hour (*ACH*). The EWNS inactivation was found to vary with the ventilation rate, which affected the contact time between EWNS and bioaerosols, and the evaporation rate of EWNS. It was also found that the nano-scale EWNS droplets showed better inactivation for smaller size fractions of bioaerosol. Thus, compared with current methods, such as HEPA filtration and oil spraying, this environmental-friendly technology (EWNS) shows promise in deactivating bioaerosols in livestock buildings. However, an in-barn study is required before its full adaption to commercial livestock barns.

4.6 Acknowledgments

Financial support from the Agriculture Development Fund (ADF) from the Ministry of Agriculture of Saskatchewan and Agrivita Canada Inc. are greatly appreciated. RLee Prokopishyn is acknowledged for his support in the design and construction of EWNS generator.

Chapter 5 – Conclusions and recommendations

5.1 Summary of results

A new designed EWNS generator was tested for both livestock barn dust reduction and bioaerosol inactivation. For the dust reduction, operating conditions including applied voltage, polarity of charge, spray liquid flow rate, electrode distance, and pH of spray liquid were optimized to achieve the highest dust reduction and the most stable performance. In conclusion, the most optimized operating condition of V = + 6.5 kV, L = 2 cm, Q = 4 µl/min, and pH = 7 had the highest total swine dust reduction of 69% and 73% at ventilation rates of 5 and 15 air changes per hour (*ACH*), respectively. The most optimized operating condition of V = + 6.5 kV, L = 2 cm, Q = 4 µl/min, and pH = 12 had the highest total poultry dust reduction of 83% at ventilation rate of 15 air changes per hour (*ACH*). The water consumption of one EWNS generator was 1.92 ml/h and EWNS generation rate was 99,840 # cm^{-3} per minute at the most optimized operating condition (V = + 6.5 kV, L = 2 cm, Q = 4 µl/min, and pH = 7). The relationships between operating parameters and water droplet size were characterized by AFM. For changing spray liquid flow rate, the smaller the flow rate, the smaller the droplets that were generated. For changing the electrode distance, as distance increased from 2 cm to 3 cm and ended till 4 cm, the randomness of the droplet size distribution was found and the droplet size became less consistent. For changing pH of sprayed liquid, as the liquid pH increased, the size of the water droplets increased.

For the *E. coli* inactivation experiments, the operating condition from the pervious study of EWNS on surface deactivation was employed, which included V = - 5.5 kV, L = 2 cm, and Q = 1 µl/min. The spray liquid of pH 7 RO water showed the highest total E. coli inactivation of 69% and 37% at ventilation rates of 7 and 15 air changes per hour (*ACH*), respectively. The size of EWNS generated from three different spray liquids were measured by AFM. It was found that pH 7 RO water and pH 12 RO water produced mean diameters of

45 and 40 nm, respectively. The droplet size of 0.9% saline could not be determined due to the interference from NaCl particles. The reactive oxygen species including hydroxy and superoxide radicals were measured by EPR. There were no discernable differences in terms of hydroxyl radical concentration between the three spray liquids. The signal of superoxide radical was not found in any of three spray liquids.

5.2 Conclusions

(1) The newly designed generator had high EWNS generation rate (99,840 # cm^{-3} per minute) with less water consumption (1.92 mL/h) than previous design of wet electrostatic scrubber for barn dusts reduction. For *E. coli* inactivation, the water consumption was only 480 µl/min.

(2) Polarity, flow rate and electrode distance were the dominant factors affecting the swine dust reduction.

(3) The e pH value of the spray liquids tends to contribute more for the poultry dust than the swine dust reduction.

(4) The best dust reduction was achieved at V= + 6.5kV, L = 2 cm, Q = 4 µL/min, pH = 7 with a total dust reduction of 72.9%.

(5) For swine dust reduction, from T-test result, there was no statistical significance between the two air exchange rates (69.0% at 5 *ACH* Vs. 72.9% at 15 *ACH*).

(6) At the treatment condition: V= + 6.5 kV, L = 2 cm, Q = 4 µL/min, pH = 12, the total reduction of poultry dust trials can reach up to 83.36%, comparing with swine dust (72.28%) at the same conditions.

(7) *E. coli* inactivation is higher when EWNS is used than that with non-thermal plasma.

(8) There were no discernable differences in terms of hydroxyl radical concentration between different treatments. Superoxide radicals can not be found in any of the treatments.

(10) *E. coli* inactivation of EWNS decreases as ventilation rate increases.

5.3 Recommendations

Overall, EWNS was shown to be an effective method for livestock fine dust reduction and bioaerosol inactivation at a lab scale level.

E. coli was used as a test agent in this study to determine the bacteria inactivation as it is a common gram-negative bacteria found in livestock barns. However, many other types of bacteria and viruses exist in livestock barns, including gram-positive bacteria *Enterococcus* and influenza viruses (e.g., H1N1), which may be differentially impacted by EWNS. Gram-positive bacteria have thicker layers of cell walls compared to the thinner layers of cell walls found in gram-negative bacteria. Moreover, the ability of EWNS to deactivate common viruses and reduce other hazardous agents such as gasses (i.e., ammonia, hydrogen sulfide) is not known. Thus, the effect of EWNS on other common contaminants in livestock operations needs to be investigated in future work.

Further, the performance of EWNS on dust reduction and bacterial deactivation at real barn conditions, which has a higher ventilation rate, larger variation of relative humidity, and larger space volume, needs to be undertaken in future studies.

References

[1] Just, N., Kirychuk, S., Gilbert, Y., Létourneau, V., Veillette, M., Singh, B., & Duchaine, C. (2011). Bacterial diversity characterization of bioaerosols from cage-housed and floor-housed poultry operations. *Environmental Research*, *111*(4), 492-498.

[2] Predicala, B. Z., Maghirang, R. G., Jerez, S. B., Urban, J. E., & Goodband, R. D. (2001). Dust and bioaerosol concentrations in two swine–finishing buildings in Kansas. *Transactions of the ASAE*, *44*(5), 1291.

[3] Rimac, D., Macan, J., Varnai, V. M., Vučemilo, M., Matković, K., Prester, L., & Pavičić, I. (2010). Exposure to poultry dust and health effects in poultry workers: impact of mould and mite allergens. *International Archives of Occupational and Environmental Health*, *83*(1), 9-19.

[4] Donham, K. (1999). A historical overview of research on the hazards of dust in livestock buildings. In *Dust Control in Animal Production Facilities, Aarhus (Denmark), 30 May-2 Jun 1999*. DJF.

[5] Borra, J. P. (2018). Review on water electro-sprays and applications of charged drops with focus on the corona-assisted cone-jet mode for High Efficiency Air Filtration by wet electro-scrubbing of aerosols. *Journal of Aerosol Science*, *125*, 208-236.

[6] Pyrgiotakis, G., McDevitt, J., Bordini, A., Diaz, E., Molina, R., Watson, C., & Demokritou, P. (2014). A chemical free, nanotechnology-based method for airborne bacterial inactivation using engineered water nanostructures. *Environmental Science: Nano*, *1*(1), 15-26.

[7] Di Natale, F., Carotenuto, C., Addio, L. D., Lancia, A., Antes, T., Szudyga, M., & Balachandran, W. (2013). New technologies for marine diesel engine emission control. *Chemical Engineering Transactions*, *32*, 361-366.

[8] Gaskell, S. J. (1997). Electrospray: principles and practice. *Journal of Mass Spectrometry*, *32*(7), 677-688.

[9] Taflin, D. C., Ward, T. L., & Davis, E. J. (1989). Electrified droplet fission and the Rayleigh limit. *Langmuir*, *5*(2), 376-384.

[10] Si, Y., Yang, Y., Martel, M., Zhang, L., Kirychuk, S., Predicala, B., & Guo, H. (2021). Characterization of electrical current and liquid droplets deposition area in a capillary electrospray. *Results in Engineering*, *9*, 100206.

[11] Si, Jordan (2020). Investigating Efficiency of Engineered Water Nanostructures (EWNS) Generated via Electrospray Technique to Deactivate Surface Microbes in Livestock Barns (Master of Science, University of Saskatchewan, Saskatoon, Canada). Retrieved from https://harvest.usask.ca/bitstream/handle/10388/12995/SI-THESIS-2018.pdf?sequence=1&isAllowed=y

[12] Wang, M., & Zhao, Q. (2019). Electrospinning and electrospray for biomedical applications. *Reference Module in Biomedical Sciences: Encyclopedia of Biomedical Engineering*.

[13] Lee, K., Lee, K. I., Jeon, S. Y., & Kim, S. (2019). Preparation of monodisperse charged droplets via electrohydrodynamic device for the removal of fine dust particles smaller than 10 μm. *Advanced Powder Technology*, *30*(1), 190-198.

[14] Jaworek, A., & Krupa, A. (1999). Jet and drops formation in electrohydrodynamic spraying of liquids. A systematic approach. *Experiments in Fluids*, *27*(1), 43-52.

[15] Kujundzic, E., Matalkah, F., Howard, C. J., Hernandez, M., & Miller, S. L. (2006). UV air cleaners and upper-room air ultraviolet germicidal irradiation for controlling airborne bacteria and fungal spores. *Journal of Occupational and Environmental Hygiene*, *3*(10), 536-546.

[16] Gañán-Calvo, A. M. (1999). The surface charge in electrospraying: its nature and its universal scaling laws. *Journal of Aerosol Science*, *30*(7), 863-872.

[17] Xie, J., Jiang, J., Davoodi, P., Srinivasan, M. P., & Wang, C. H. (2015). Electrohydrodynamic atomization: A two-decade effort to produce and process micro-nanoparticulate materials. *Chemical Engineering Science*, *125*, 32-57.

[18] Hartman, R. P. A., Brunner, D. J., Camelot, D. M. A., Marijnissen, J. C. M., & Scarlett, B. (1999). Electrohydrodynamic atomization in the cone–jet mode physical modeling of the liquid cone and jet. *Journal of Aerosol Science*, *30*(7), 823-849.

[19] Hamon, L., Andrès, Y., & Dumont, E. (2012). Aerial pollutants in swine buildings: A review of their characterization and methods to reduce them. *Environmental Science & Technology*, *46*(22), 12287-12301.

[20] Donham, K. J. (1986). Hazardous agents in agricultural dusts and methods of evaluation. *American Journal of Industrial Medicine*, *10*(3), 205-220.

[21] Zhang, Y., Nijssen, L., Barber, E. M., Feddes, J. J., & Sheridan, M. (1994). Sprinkling mineral oil to reduce dust concentration in swine buildings. *ASHRAE Transactions*, *100*(2), 1043-1050.

[22] Zhang, Y., Tanaka, A., Dosman, J. A., Senthilselvan, A., Barber, E. M., Kirychuk, S. P., & Hurst, T. S. (1998). Acute respiratory responses of human subjects to air quality in a swine building. *Journal of Agricultural Engineering Research*, *70*(4), 367-373.

[23] Almuhanna, E. A., Maghirang, R. G., Murphy, J. P., & Erickson, L. E. (2009). Laboratory scale electrostatically assisted wet scrubber for controlling dust in livestock buildings. *Applied Engineering in Agriculture*, *25*(5), 745-750.

[24] Lai, K. M., Burge, H. A., & First, M. W. (2004). Size and UV germicidal irradiation susceptibility of Serratia marcescens when aerosolized from different suspending media. *Applied and Environmental Microbiology*, *70*(4), 2021-2027.

[25] Mead, K., & Johnson, D. L. (2004). An evaluation of portable high-efficiency particulate air filtration for expedient patient isolation in epidemic and emergency response. *Annals of Emergency Medicine*, *44*(6), 635-645.

[26] Chen, C. Y., Wu, L. C., Chen, H. Y., & Chung, Y. C. (2010). Inactivation of Staphylococcus aureus and Escherichia coli in water using photocatalysis with fixed TiO 2. *Water, Air, & Soil Pollution*, *212*(1), 231-238.

[27] Liao, X., Liu, D., Xiang, Q., Ahn, J., Chen, S., Ye, X., & Ding, T. (2017). Inactivation mechanisms of non-thermal plasma on microbes: A Review. *Food Control*, *75*, 83-91.

[28] Zaffina, S., Camisa, V., Lembo, M., Vinci, M. R., Tucci, M. G., Borra, M., ... & Cannatà, V. (2012). Accidental exposure to UV radiation produced by germicidal lamp: case report and risk assessment. *Photochemistry and Photobiology*, *88*(4), 1001-1004.

[29] Kligman, L. H., Akin, F. J., & Kligman, A. M. (1985). The contributions of UVA and UVB to connective tissue damage in hairless mice. *Journal of Investigative Dermatology*, *84*(4).

[30] Adal, K. A., Anglim, A. M., Palumbo, C. L., Titus, M. G., Coyner, B. J., & Farr, B. M. (1994). The use of high-efficiency particulate air-filter respirators to protect hospital workers from tuberculosis--a cost-effectiveness analysis. *New England Journal of Medicine*, *331*(3), 169-173.

[31] Viral Penetration of High Particulate Air (HEPA) Filters (2007). In *Defense Technical Information Centre*. Retrieved August 25, 2019, from apps.dtic.mil/docs/citations/ADA464232

[32] Gupta, S. M., & Tripathi, M. (2012). An overview of commonly used semiconductor nanoparticles in photocatalysis. *High Energy Chemistry*, *46*(1), 1-9.

[33] Sotiriou, G. A., Diaz, E., Long, M. S., Godleski, J., Brain, J., Pratsinis, S. E., & Demokritou, P. (2012). A novel platform for pulmonary and cardiovascular toxicological characterization of inhaled engineered nanomaterials. *Nanotoxicology*, *6*(6), 680-690.

[34] Hsiao, I. L., & Huang, Y. J. (2011). Effects of various physicochemical characteristics on the toxicities of ZnO and TiO2 nanoparticles toward human lung epithelial cells. *Science of the Total Environment*, *409*(7), 1219-1228.

[35] Jaworek, A., Balachandran, W., Lackowski, M., Kulon, J., & Krupa, A. (2006). Multi-nozzle electrospray system for gas cleaning processes. *Journal of Electrostatics*, *64*(3-4), 194-202.

[36] Unger, L., Ehouarn, P., & Borra, J. P. (2003). Filtration of submicron sized particles by electrocoagulation. *Journal of Aerosol Science*, *34*, 501-502.

[37] Wang, B., Yin, J. J., Zhou, X., Kurash, I., Chai, Z., Zhao, Y., & Feng, W. (2013). Physicochemical origin for free radical generation of iron oxide nanoparticles in biomicroenvironment: catalytic activities mediated by surface chemical states. *The Journal of Physical Chemistry C*, *117*(1), 383-392.

[38] Jaworek, A., Adamiak, K., Balachandran, W., Krupa, A., Castle, P., & Machowski, W. (2002). Numerical simulation of scavenging of small particles by charged droplets. *Aerosol Science & Technology*, *36*(9), 913-924.

[39] Krupa, A., Jaworek, A., Szudyga, M., Czech, T., Sobczyk, A. T., Marchewicz, A., & D'Addio, L. (2016). Diesel nanoparticles removal by charged spray. *International Journal of Plasma Environmental Science and Technology*, *10*(2).

[40] Stadtman, E. R. (1992). Protein oxidation and aging. *Science*, *257*(5074), 1220-1224.

[41] Imlay, J. A., & Linn, S. (1988). DNA damage and oxygen radical toxicity. *Science*, *240*(4857), 1302-1309.

[42] Jeong, J., Kim, J. Y., & Yoon, J. (2006). The role of reactive oxygen species in the electrochemical inactivation of microorganisms. *Environmental Science & Technology*, *40*(19), 6117-6122.

[43] Cabiscol Català, E., Tamarit Sumalla, J., & Ros Salvador, J. (2000). Oxidative stress in bacteria and protein damage by reactive oxygen species. *International Microbiology, 2000,* vol. 3, num. 1, p. 3-8.

[44] Humphries, K. M., & Szweda, L. I. (1998). Selective inactivation of α-ketoglutarate dehydrogenase and pyruvate dehydrogenase: reaction of lipoic acid with 4-hydroxy-2-nonenal. *Biochemistry*, *37*(45), 15835-15841.

[45] He, W., Zhou, Y. T., Wamer, W. G., Boudreau, M. D., & Yin, J. J. (2012). Mechanisms of the pH dependent generation of hydroxyl radicals and oxygen induced by Ag nanoparticles. *Biomaterials*, *33*(30), 7547-7555.

[46] He, W., Zhou, Y. T., Wamer, W. G., Hu, X., Wu, X., Zheng, Z., ... & Yin, J. J. (2013). Intrinsic catalytic activity of Au nanoparticles with respect to hydrogen peroxide decomposition and superoxide scavenging. *Biomaterials*, *34*(3), 765-773.

[47] Zhang, W., Li, Y., Niu, J., & Chen, Y. (2013). Photogeneration of reactive oxygen species on uncoated silver, gold, nickel, and silicon nanoparticles and their antibacterial effects. *Langmuir*, *29*(15), 4647-4651.

[48] Vankayala, R., Kuo, C. L., Sagadevan, A., Chen, P. H., Chiang, C. S., & Hwang, K. C. (2013). Morphology dependent photosensitization and formation of singlet oxygen (1 Δ g) by gold and silver nanoparticles and its application in cancer treatment. *Journal of Materials Chemistry B*, *1*(35), 4379-4387.

[49] Yin, J. J., Liu, J., Ehrenshaft, M., Roberts, J. E., Fu, P. P., Mason, R. P., & Zhao, B. (2012). Phototoxicity of nano titanium dioxides in HaCaT keratinocytes—generation of reactive oxygen species and cell damage. *Toxicology and Applied Pharmacology*, *263*(1), 81-88.

[50] Tao, F., Gonzalez-Flecha, B., & Kobzik, L. (2003). Reactive oxygen species in pulmonary inflammation by ambient particulates. *Free Radical Biology and Medicine*, *35*(4), 327-340.

[51] Haegens, A., van der Vliet, A., Butnor, K. J., Heintz, N., Taatjes, D., Hemenway, D., ... & Mossman, B. T. (2005). Asbestos-induced lung inflammation and epithelial cell proliferation are altered in myeloperoxidase-null mice. *Cancer Research*, *65*(21), 9670-9677.

[52] Rhoden, C. R., Lawrence, J., Godleski, J. J., & González-Flecha, B. (2004). N-acetylcysteine prevents lung inflammation after short-term inhalation exposure to concentrated ambient particles. *Toxicological Sciences*, *79*(2), 296-303.

[53] Pyrgiotakis, G., McDevitt, J., Brain, J., & Demokritou, P. (2020, May 8). A novel method for bacterial inactivation using engineered water nanostructures. In *Center for Nanotechnology and Nanotoxicology*. Retrieved from http://hsph.harvard.edu/nano

[54] Thorne, P. S. (2007). Environmental health impacts of concentrated animal feeding operations: anticipating hazards—searching for solutions. *Environmental Health Perspectives*, *115*(2), 296-297.

[55] Aarnink, A. J. A., & Ellen, H. H. (2007). Processes and factors affecting dust emissions from livestock production. *How to Improve Air Quality*.

[56] Criteria air contaminants—2006 Emission summaries (2008). In *Environment Canada*. Retrieved February 25, 2020, from http://www.ec.gc.ca/pdb/cac/Emissions1990-2015/emissions_e.cfm

[57] Pattey, E., & Qiu, G. (2012). Trends in primary particulate matter emissions from Canadian agriculture. *Journal of the Air & Waste Management Association*, *62*(7), 737-747.

[58] Mostafa, E., & Buescher, W. (2011). Indoor air quality improvement from particle matters for laying hen poultry houses. *Biosystems Engineering*, *109*(1), 22-36.

[59] Winkel, A., Mosquera, J., Aarnink, A. J., Koerkamp, P. W. G., & Ogink, N. W. (2016). Evaluation of oil spraying systems and air ionisation systems for abatement of particulate matter emission in commercial poultry houses. *Biosystems Engineering*, *150*, 104-122.

[60] Winkel, A., Mosquera, J., Aarnink, A. J., Koerkamp, P. W. G., & Ogink, N. W. (2015). Evaluation of a dry filter and an electrostatic precipitator for exhaust air cleaning at commercial non-cage laying hen houses. *Biosystems Engineering*, *129*, 212-225.

[61] Poppendieck, D. G., Rim, D., & Persily, A. K. (2014). Ultrafine particle removal and ozone generation by in-duct electrostatic precipitators. *Environmental Science & Technology*, *48*(3), 2067-2074.

[62] Kiss, I., Iváncsy, T., Németh, B., & Berta, I. (2009). Advanced risk analysis for the application of ESP-s to clean flammable gas-pollutant mixtures. In *Electrostatic Precipitation* (pp. 50-53). Springer, Berlin, Heidelberg.

[63] Jaworek, A., Krupa, A., & Czech, T. (2007). Modern electrostatic devices and methods for exhaust gas cleaning: A brief review. *Journal of Electrostatics*, *65*(3), 133-155.

[64] Di Natale, F., Carotenuto, C., D'Addio, L., Jaworek, A., Krupa, A., Szudyga, M., & Lancia, A. (2015). Capture of fine and ultrafine particles in a wet electrostatic scrubber. *Journal of Environmental Chemical Engineering*, *3*(1), 349-356.

[65] Krupa, A., Jaworek, A., Sobczyk, A. T., Marchewicz, A., Szudyga, M., & Antes, T. (2013). Charged spray generation for gas cleaning applications. *Journal of Electrostatics*, *71*(3), 260-264.

[66] Kim, H. G., Kim, H. J., Lee, M. H., & Kim, J. H. (2014). Experimental study on the enhancement of particle removal efficiency in spray tower scrubber using electrospray. *Asian Journal of Atmospheric Environment*, *8*(2), 89-95.

[67] Gañán-Calvo, A. M. (2004). On the general scaling theory for electrospraying. *Journal of Fluid Mechanics, 507, 203-212a.*

[68] Zhu, L., Ding, X., & Wu, X. (2020). A novel method for improving the anti-pilling property of knitted wool fabric with engineered water nanostructures. *Journal of Materials Research and Technology*, *9*(3), 3649-3658.

[69] Zhu, L., Liu, Y., Ding, X., Wu, X., Sand, W., & Zhou, H. (2019). A novel method for textile odor removal using engineered water nanostructures. *RSC Advances*, *9*(31), 17726-17736.

[70] Wang, P. K. (1986). Brownian diffusion of charged fine particles surrounding a conducting cylinder in the presence of an external electric field. *Journal of Aerosol Science*, *17*(2), 201-209.

[71] SCoEaNIHR, E. S. (2010). Scientific Basis for the Definition of the Term "nanomaterial". *European Commission, Scientific Committee on Emerging and Newly Identified Health Risks (SCENHR)*.

[72] Heating, ventilation, and air conditioning: Addendum to the CCAC guidelines on laboratory animal facilities – characteristics, design and development (n.d.). In *Canadian Council on Animal Care*. Retrieved January 25, 2021, from www.ccac.ca/Documents/Standards/Guidelines/Addendum-Laboratory-Animals-Facilities_heating-ventilation-air-conditioning.pdf. Accessed 5 Jan. 2021

[73] Pilote, J., Létourneau, V., Girard, M., & Duchaine, C. (2019). Quantification of airborne dust, endotoxins, human pathogens and antibiotic and metal resistance genes in Eastern Canadian swine confinement buildings. *Aerobiologia*, *35*(2), 283-296.

[74] Cherrie, J. W., Brosseau, L. M., Hay, A., & Donaldson, K. (2013). Low-toxicity dusts: current exposure guidelines are not sufficiently protective. *Annals of Occupational Hygiene*, *57*(6), 685-691.

[75] Ganan-Calvo, A. M., Davila, J., & Barrero, A. (1997). Current and droplet size in the electrospraying of liquids. Scaling laws. *Journal of Aerosol Science*, *28*(2), 249-275.

[76] Seto, T., Maekawa, T., Osone, S., Kawamura, K., Yamauchi, T., & Otani, Y. (2013). Formation of highly charged nanodroplets by condensation–electrospray device. *Chemical Engineering Science*, *85*, 46-49.

[77] Vaze, N., Pyrgiotakis, G., Mena, L., Baumann, R., Demokritou, A., Ericsson, M., & Demokritou, P. (2019). A nano-carrier platform for the targeted delivery of nature-inspired antimicrobials using Engineered Water Nanostructures for food safety applications. *Food Control*, *96*, 365-374.

[78] Bruins, A. P. (1998). Mechanistic aspects of electrospray ionization. *Journal of Chromatography A*, *794*(1-2), 345-357.

[79] Ikonomou, M. G., Blades, A. T., & Kebarle, P. (1990). Investigations of the electrospray interface for liquid chromatography/mass spectrometry. *Analytical Chemistry*, *62*(9), 957-967.

[80] Teng, C., & Li, J. (2020). Experimental Study on Particle Removal of a Wet Electrostatic Precipitator with Atomization of Charged Water Drops. *Energy & Fuels*, *34*(6), 7257-7268.

[81] Aarnink, A. J. A., Roelofs, P. F. M. M., Ellen, H., & Gunnink, H. (1999). Dust sources in animal houses. In *Proceedings of the International Symposium on Dust Control in Animal Production Facilities, 30 May-2 June, Aarhus (D)* (pp. 34-40).

[82] Takai, H., & Pedersen, S. (2002). Livestock related fine dust–composition, structure and flows. *Landbauforschung Völkenrode*, *235*, 139-144.

[83] Canadian Live Swine and Pork Exports (n.d.). In *Canadian Pork Council*. Retrieved January 4, 2021, from http://www.cpc-ccp.com/canadian-pork-and-live-swine-exports

[84] Brisson, Y. (2014). The changing face of the Canadian hog industry.

[85] Walker, J. T., Aneja, V. P., & Dickey, D. A. (2000). Atmospheric transport and wet deposition of ammonium in North Carolina. *Atmospheric Environment*, *34*(20), 3407-3418.

[86] Hedelin, A. S., Sundblad, B. M., Sahlander, K., Wilkinson, K., Seisenbaeva, G., Kessler, V., & Palmberg, L. (2016). Comparing human respiratory adverse effects after acute exposure to particulate matter in conventional and particle-reduced swine building environments. *Occupational and Environmental Medicine, 73*(10), 648-655.

[87] Iversen, M., Kirychuk, S., Drost, H., & Jacobson, L. (2000). Human health effects of dust exposure in animal confinement buildings. *Journal of Agricultural Safety and Health, 6*(4), 283.

[88] Keessen, E. C., Harmanus, C., & Dohmen, W. (2013). Clostridium difficile infection associated with pig farms. *Emerging Infectious Diseases, 19*(6), 1032.

[89] Poggenborg, R., Gaïni, S., Kjaeldgaard, P., & Jørgen Christensen, J. (2008). Streptococcus suis: meningitis, spondylodiscitis and bacteraemia with a serotype 14 strain. *Scandinavian Journal of Infectious Diseases*, *40*(4), 346-349.

[90] Vaze, N., Pyrgiotakis, G., McDevitt, J., Mena, L., Melo, A., Bedugnis, A., & Demokritou, P. (2019). Inactivation of common hospital acquired pathogens on surfaces and in air utilizing engineered water nanostructures (EWNS) based nano-sanitizers. *Nanomedicine: Nanotechnology, Biology and Medicine*, *18*, 234-242.

[91] Pyrgiotakis, G., Vedantam, P., Cirenza, C., McDevitt, J., Eleftheriadou, M., Leonard, S. S., & Demokritou, P. (2016). Optimization of a nanotechnology based antimicrobial platform for food safety applications using Engineered Water Nanostructures (EWNS). *Scientific Reports*, *6*(1), 1-12.

[92] Huang, R., Vaze, N., Soorneedi, A., Moore, M. D., Xue, Y., Bello, D., & Demokritou, P. (2019). Inactivation of Hand Hygiene-Related Pathogens Using Engineered Water Nanostructures. *ACS Sustainable Chemistry & Engineering*, *7*(24), 19761-19769.

[93] Lee, J. H., & Jo, W. K. (2006). Characteristics of indoor and outdoor bioaerosols at Korean high-rise apartment buildings. *Environmental Research*, *101*(1), 11-17.

[94] Mandal, J., & Brandl, H. (2011). Bioaerosols in indoor environment-a review with special reference to residential and occupational locations. *The Open Environmental & Biological Monitoring Journal*, *4*(1).

[95] Létourneau, V., Nehmé, B., Mériaux, A., Massé, D., Cormier, Y., & Duchaine, C. (2010). Human pathogens and tetracycline-resistant bacteria in bioaerosols of swine confinement buildings and in nasal flora of hog producers. *International Journal of Hygiene and Environmental Health*, *213*(6), 444-449.

[96] Thorne, P. S., Ansley, A. C., & Perry, S. S. (2009). Concentrations of bioaerosols, odors, and hydrogen sulfide inside and downwind from two types of swine livestock operations. *Journal of Occupational and Environmental Hygiene*, *6*(4), 211-220.

[97] Tresp, H., Hammer, M. U., Winter, J., Weltmann, K. D., & Reuter, S. (2013). Quantitative detection of plasma-generated radicals in liquids by electron paramagnetic resonance spectroscopy. *Journal of Physics D: Applied Physics*, *46*(43), 435401.

[98] Sun, P., Sun, Y., Wu, H., Zhu, W., Lopez, J. L., Liu, W., ... & Fang, J. (2011). Atmospheric pressure cold plasma as an antifungal therapy. *Applied Physics Letters*, *98*(2), 021501.

[99] Tani, A., Ono, Y., Fukui, S., Ikawa, S., & Kitano, K. (2012). Free radicals induced in aqueous solution by non-contact atmospheric-pressure cold plasma. *Applied Physics Letters*, *100*(25), 254103.

[100] Lee, Y. H., & Lee, B. U. (2006). Inactivation of airborne E. coli and B. subtilis bioaerosols utilizing thermal energy. *Journal of Microbiology and Biotechnology*, *16*(11), 1684-1689.

[101] Pal, A., Pehkonen, S. O., Yu, L. E., & Ray, M. B. (2008). Photocatalytic inactivation of airborne bacteria in a continuous-flow reactor. *Industrial & Engineering Chemistry Research*, *47*(20), 7580-7585.

[102] Macher, J. M. (1989). Positive-hole correction of multiple-jet impactors for collecting viable microorganisms. *American Industrial Hygiene Association Journal*, *50*(11), 561-568.

[103] Tan, Z., & Zhang, Y. (2004). A review of effects and control methods of particulate matter in animal indoor environments. *Journal of the Air & Waste Management Association*, *54*(7), 845-854.

[104] Rayleigh, L. (1882). On the equilibrium of liquid conducting masses charged with electricity. *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science*, *14*(87), 184-186.

[105] Thomas, R. J., Webber, D., Sellors, W., Collinge, A., Frost, A., Stagg, A. J., ... & Titball, R. W. (2008). Characterization and deposition of respirable large-and small-particle bioaerosols. *Applied and Environmental Microbiology*, *74*(20), 6437-6443.

[106] Wysok, B., Uradziński, J., & Gomólka-Pawlicka, M. (2006). Ozone as an alternative disinfectant-a review. *Polish Journal of Food and Nutrition Sciences*, *15*(1), 3.

Appendix A – Experiment setup

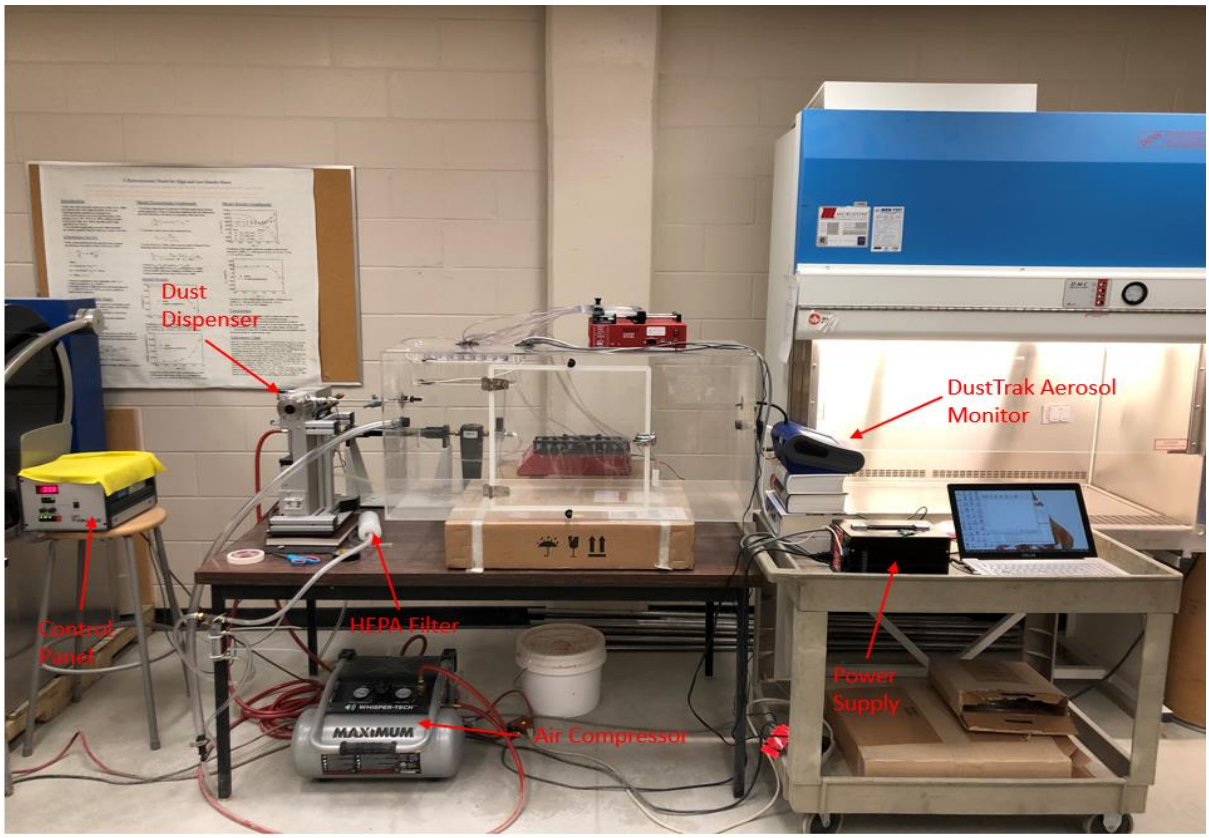

Figure A.1 Experiment setup of dust reduction

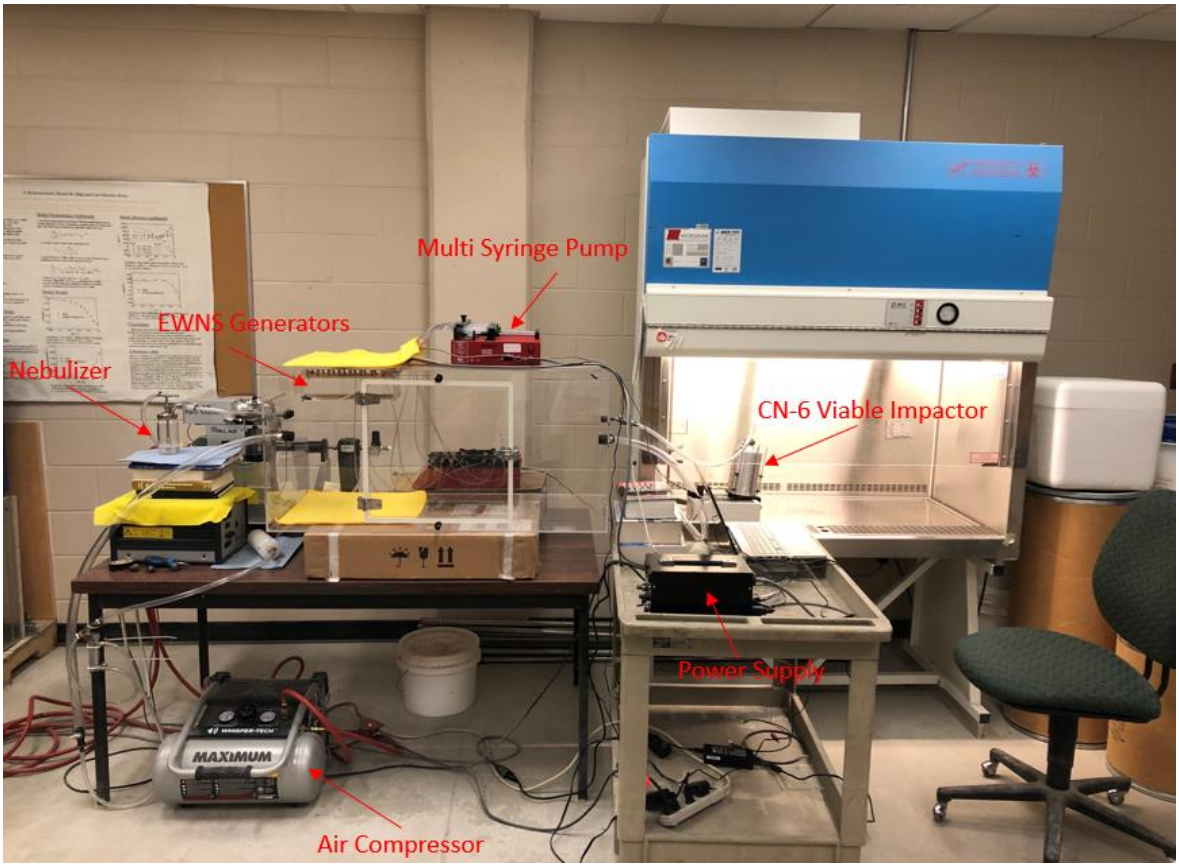

Figure A.2 Experiment setup of *E. coli* inactivation

Appendix B Procedures of preparing dust samples

B.1) Collection of livestock barn dusts

a. The settled swine dust samples were collected from the top of the penning in six rooms at the swine barn facility of the Prairie Swine Centre.

b. The settle poultry dust samples were collected from the floor in different broiler rooms at the Poultry centre at University of Saskatchewan.

B.2) Preparation of dust cake

a. The settled swine and poultry dust were sieved by a 100 μm sieve to remove large particles (feather, faeces, and feed).

b. The sieved dust samples were sieved into smaller size fractions by a 50 μm sieve.

c. Dry the sieved dust samples by using the oven at 100 °C for 4 hours and ensure the moisture content of each dust sample is below 2% by using moisture content analyzer.

d. Feed the dried dust samples into the dust dispenser column and use pestle to press the dust sample to form a dust cake.

Appendix C Procedures of preparing *E. coli* bioaerosol

C.1) Preparation of LB broth

a. Weigh out 12.5g of LB Broth powder (BD Difco, Fisher Scientific, Cat#DF0446-17-3) using a top loading balance and place into a 1L Nalgene bottle.
b. Dissolve the LB broth powder in 500 mL of distilled water.
c. Unscrew the lid to the bottle so it is loose, cover the lid with tin foil and put a piece of autoclave tape with: media type, date, lab #, and initials/PI's initials. Put the bottles of media in a solid container and place on the cart for autoclaving.

C.2) Preparation of LB Agar plates

a. Weigh out 7.5 g of granulated Agar (BD Difco, Fisher Scientific) and 12.5g of LB broth powder using a top loading balance. Place both into a 1L Nalgene bottle.
b. Dissolve the powder in 500 mL of distilled water.
c. Unscrew the lid to the bottle so it is loose, cover the lid with tin foil and put a piece of autoclave tape with: media type, date, lab #, and initials/PI's initials. Put the bottles of media in a solid container and place on the cart for autoclaving.
d. Cool the agar down in a 65°C water bath.
e. In the biosafety cabinet, pour agar into petri plates covering approximately $\frac{3}{4}$ of the plate. Close the lids to the plates and swirl gently to distribute the agar across the petri plate.
f. Allow the plates to solidify. Store at 4°C if not using right away.

C.3) Preparation of *E. coli* after receiving from supplier

a. In the biosafety cabinet, open the vial according to the manufacturer's instructions.
b. Add 0.5 to 1 mL of LB broth to the tube containing the pellet using a pipette.
c. Mix contents of the tube by pipetting up and down gently.
d. Aseptically transfer this aliquot to a tube containing LB Broth (5 to 6 mL) and mix well.
e. Using a sterile disposable loop, streak the culture onto LB agar plates.
f. Incubate the tube and plates at 37°C for 16-18 hours.

C.4) *E. coli* inoculation (plate to plate)

a. Use sterile disposable loop to attach one single colony of *E. coli* from original agar plate.
b. Close the original petri dish.
c. Partially lift the lid of the petri dish containing the solid LB-A medium.
d. Hold the charged loop parallel with the surface of the agar, smear the inoculum backwards and forwards across a small area of the medium (see streaked area A in photograph).
e. Turn the dish 90° anticlockwise, with the same loop streak the plate from area A across the surface of the agar in three or four parallel lines (B). Make sure that a small amount of culture is carried over.
f. Turn the dish 90° anticlockwise again and streak from B across the surface of agar in three or four parallel lines (C).
g. Turn the dish 90° anticlockwise and streak loop across the surface of agar from C into the centre of the plate (D).
h. Remove and dispose the loop, close the Petri dish.
i. Seal and store at 4° for maximum two weeks.

(**All procedures of inoculation above are performed in the biosafety cabinet**.)

C.5) *E. coli* inoculation (plate to liquid culture medium):

a. Use sterile disposable loop to attach one single colony of *E. coli* from agar plate.
b. Close the Petri dish.
c. Dip the loop inside a 10 ml Falcon tube containing 5 ml of liquid LB medium and mix slowly.
d. Close the Falcon tube and dispose the loop.
e. Incubate the culture medium at 37 °C for 12 to 20 hours.

(**All procedures of inoculation above are performed in the biosafety cabinet**.)

C.6) Optical density determination

a. Take 1 mL of culture and put it into a plastic cuvette.
b. Recap the Falcon tube with liquid culture medium.
c. Wipe down the outside of the cuvette with a kimwipe to remove any thing that could interfere with the OD reading.

d. Use the single wavelength mode of UV-Vis spectrophotometer to measure the absorbance of 1 ml incubated LB medium at 600 nm (LB culture as blank).
e. If the OD>1, the sample dilution is needed.

(The OD reading can be performed on the bench)

C.7) Preparation of *E. coli* solution for aerosolization

a. Optimize the stock solution to get the optical density of 1.
b. Perform a five-hundred-fold dilution using autoclaved deionized water for the stock solution.
c. Load 80 ml of diluted stock solution into the jar of the nebulizer.

Appendix D Preparation of EPR samples

D.1) Electrospray set up

a. Prepare a series of 2.5 ml spray solution with different pH (range from 4 – 11).
b. Load the spray solution into a 12 ml plastic syringe and set up the electrospray unit.
c. Set the syringe pump flow rate and supply voltage to pre-determined values and test the spray mode to ensure it is stable.

D.2) Spin trapping agent preparation

a. Dissolve 30 µL of DMPO in 1 ml of distilled water and 1 ml ethanol respectively to detect the existence of OH · and $O_2^{\cdot -}$.
b. Pour the 1 ml of DMPO solution into a 5 ml centrifuge tube and place below the electrospray unit for a period of 20 min, only one ES unit will be needed at a time.

D.3) Optimized EPR parameters and measurement

a. X-band EPR spectroscope with a frequency of 9.75 GHz will be used, modulation frequency of 100 kHz; modulation amplitude of 0.1 mT; microwave power of 5.024 mW; receiver gain of 30 dB; time constant of 0.01 ms, and magnetic field scan of 10 mT.
b. Place 50 µL of freshly prepared trapping solution into the borosilicate glass sample tube (125 mm length, 0.8525 mm inner diameter)
c. Place the tube into the EPR to obtain a signal of the untreated spin trap solution.
d. Place 50 µL of the electro spray treated sample into a sample tube, place in the EPR and measure.
e. From the end of the treatment to the beginning of the EPR measurement a few minutes are required for handling, this delay time is held constant at exactly 4 min for all the EPR measurements.
f. When the EPR spectrum of the treated sample was measured, the spectrum of the untreated control is subtracted.
g. The relative intensity between different treatments will be compared to determine which treatment has the highest relative spin concentration.

Appendix E AFM results of EWNS

This section contains original data of droplet size at each condition in Table 3.1 from Chapter 3 - Experimental Studies on Removing Airborne Fine Dust Particles using Engineered Water Nanostructures Generated from an Electrospray.

Table E.1 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 1.

#	Height (nm)	Diameter (nm)	Equivalent diameter of droplet (nm)
1	11.89	163	98
2	9.24	137	80
3	9.47	150	86
4	9.00	150	85
5	10.58	156	92
6	4.62	130	62
7	6.84	124	68
8	8.69	143	81
9	11.37	143	89
10	5.37	130	65
11	7.61	138	76
12	5.66	144	71
13	5.31	130	65
14	7.93	144	79
15	8.97	169	92
16	2.78	107	46
17	4.56	91	49
18	12.35	165	100
19	8.21	189	96
20	11.30	156	94

Table E.2 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 2.

#	Height (nm)	Diameter (nm)	Equivalent diameter of droplet (nm)
1	0.16	151	22
2	0.07	114	14
3	0.14	165	22
4	0.15	132	20
5	0.12	128	18
6	0.05	141	14
7	0.12	92	15
8	0.12	81	13
9	0.08	81	12
10	0.05	92	11
11	0.11	117	17
12	0.05	118	13
13	0.12	153	20
14	0.12	124	18
15	0.05	162	16
16	0.12	210	25
17	0.11	177	22
18	0.12	156	21
19	0.14	130	19
20	0.12	107	16

Table E.3 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 3.

#	Height (nm)	Diameter (nm)	Equivalent diameter of droplet (nm)
1	7.73	78	52
2	4.74	73	42
3	7.57	73	50
4	5.14	70	42
5	6.81	73	48
6	8.04	74	51
7	7.02	74	49
8	5.75	59	39
9	4.51	70	40
10	5.24	60	39
11	7.10	84	53
12	6.24	73	46
13	10.71	79	59
14	5.60	73	45
15	13.45	98	73
16	3.75	78	41
17	4.78	64	39
18	4.78	78	44
19	3.04	74	37
20	1.92	36	19

Table E.4 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 4.

#	Height (nm)	Diameter (nm)	Equivalent diameter of droplet nm)
1	45.91	373	268
2	35.54	419	266
3	53.40	419	305
4	38.05	385	257
5	55.86	383	292
6	43.56	341	248
7	39.88	264	203
8	45.85	345	254
9	45.11	459	306
10	53.89	387	290
11	35.27	542	315
12	31.68	394	246
13	42.20	342	246
14	47.20	383	276
15	24.62	465	252
16	20.38	550	264
17	35.40	382	250
18	37.24	334	232
19	29.82	583	312
20	37.25	485	298

Table E.5 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 5.

#	Height (nm)	Diameter (nm)	Equivalent diameter of droplet (nm)
1	55.64	650	413
2	22.48	640	302
3	55.81	570	379
4	50.90	325	253
5	9.54	615	221
6	25.70	563	290
7	3.24	369	110
8	43.17	260	207
9	77.20	287	269
10	10.41	721	253
11	32.27	329	219
12	19.18	749	318
13	29.45	374	231
14	13.97	385	184
15	32.63	533	303
16	46.75	575	360
17	22.16	423	228
18	13.26	365	174
19	27.99	543	292
20	24.63	483	258

Table E.6 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 6.

#	Height (nm)	Diameter (nm)	Equivalent diameter of droplet (nm)
1	7.73	78	52
2	4.74	73	42
3	7.57	73	50
4	5.14	70	42
5	6.81	73	48
6	8.04	74	51
7	7.02	74	49
8	5.75	59	39
9	4.51	70	40
10	5.24	60	39
11	7.10	84	53
12	6.24	73	46
13	10.71	79	59
14	5.60	73	45
15	13.45	98	73
16	3.75	78	41
17	4.78	64	39
18	4.78	78	44
19	3.04	74	37
20	1.92	36	19

Table E.7 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 7.

#	Height (nm)	Diameter (nm)	Equavilent diameter of droplet (nm)
1	0.52	65	19
2	0.51	60	18
3	0.66	50	17
4	0.71	65	21
5	0.38	48	14
6	0.60	60	19
7	0.21	51	12
8	0.33	53	14
9	0.63	42	15
10	0.44	55	16
11	0.29	53	13
12	0.48	56	16
13	0.75	45	17
14	0.36	50	14
15	0.43	45	14
16	0.28	55	14
17	0.54	55	17
18	0.39	73	18
19	0.62	56	18
20	0.45	64	18

Table E.8 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 8.

#	Height (nm)	Diameter (nm)	Equivalent diameter of droplet nm)
1	5.99	73	46
2	3.31	38	24
3	3.79	50	30
4	1.85	55	26
5	3.57	42	27
6	4.10	50	31
7	4.70	59	36
8	1.23	46	20
9	1.17	37	17
10	1.43	39	19
11	1.37	57	24
12	0.98	35	15
13	1.09	49	20
14	1.25	42	19
15	1.34	57	24
16	1.23	49	21
17	1.82	54	25
18	2.49	62	31
19	1.09	52	21
20	3.50	39	25

Table E.9 The original data of the diameter a and the height h of the spread droplets to calculate the equivalent diameter of EWNS at condition 9.

#	Height (nm)	Diameter (nm)	Equivalent diameter of droplet (nm)
1	4.75	158	71
2	4.08	137	61
3	4.56	108	54
4	17.63	120	91
5	9.29	127	77
6	13.94	134	91
7	10.75	100	69
8	14.60	150	100
9	11.71	143	90
10	5.94	105	58
11	13.41	108	78
12	7.95	133	75
13	4.56	71	41
14	1.49	84	32
15	2.62	81	37
16	0.75	65	21
17	9.11	79	56
18	2.68	78	37
19	0.42	79	20
20	0.83	78	25

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Editor of portion(s)	N/A	Issue, if republishing an article from a serial	N/A
Volume of serial or monograph	N/A	Publication date of portion	2013-12-31
Page or page range of portion	14-17		

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